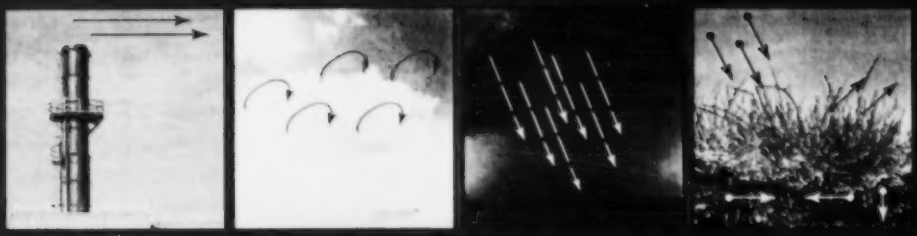
Canada's Nuclear Regulator

Investigation of the Environmental Fate of Tritium in the Atmosphere

Part of the Tritium Studies Project

INFO-0792



Report prepared for the CNSC by EcoMetrix Incorporated in association with RWDI Air Inc.

December 2009

Canadian Nuclear Safety Commission
Commission canadienne de sûreté nucléaire

Canada

Investigation of the Environmental Fate of Tritium in the Atmosphere

© Minister of Public Works and Government Services Canada 2009
Catalogue number CC172-51/2009E-PDF
ISBN 978-1-100-13928-9

Published by the Canadian Nuclear Safety Commission (CNSC)
Catalogue number: INFO-0792

Report prepared for the CNSC by ECOMETRIX Incorporated in association with RWDI Air Inc. (RSP-0247), March 2009.

Extracts from this document may be reproduced for individual use without permission provided the source is fully acknowledged. However, reproduction in whole or in part for purposes of resale or redistribution requires prior written permission from the Canadian Nuclear Safety Commission.

Également publié en français sous le titre de : *Le devenir environnemental du tritium dans l'atmosphère*

Document availability
This document can be viewed on the CNSC Web site at nuclearsafety.gc.ca. To order a printed copy of the document in English or French, please contact:

Canadian Nuclear Safety Commission
280 Slater Street
P.O. Box 1046, Station B
Ottawa, Ontario K1P 5S9
CANADA

Tel.: 613-995-5894 or 1-800-668-5284 (in Canada only)
Facsimile: 613-995-5086
E-mail: info@cnsc-ccsn.gc.ca
Web site: nuclearsafety.gc.ca

Cover images
The cover images depict the tritium cycle in the atmosphere. Tritium is released from the stack and disperses in the air. It is then deposited on vegetation and soil by rainfall (washout) or through vapour exchange. The tritium may then be re-emitted from the plants and soil to the atmosphere.

TABLE OF CONTENTS

EXECUTIVE SUMMARY 1

1.0 INTRODUCTION 3

2.0 SOURCES AND CHEMICAL FORMS OF TRITIUM 4
2.1 Chemical Forms 4
2.2 Natural Abundance 4
2.3 Natural Sources 5
2.4 Anthropogenic Sources 7
2.4.1 Thermonuclear Detonation during Nuclear Weapons Testing 7
2.4.2 Nuclear Reactors 11
2.4.2.1 Pressurized Water Reactors (PWRs) 13
2.4.2.2 Boiling Water Reactors (BWRs) 16
2.4.2.3 Heavy Water Reactors (HWRs) 17
2.4.2.4 Gas-Cooled Reactors (GCRs) 18
2.4.2.5 Airborne Tritium Releases from Reactors 19
2.4.2.6 Liquid Tritium Releases from Reactors 19
2.4.2.7 Controlled Thermonuclear (or Fusion) Reactors 19
2.4.2.8 Influence of Cooling Water Systems on Tritium Release 20
2.4.3 Fuel Reprocessing Plants 21
2.4.4 Tritium Production Facilities 21
2.4.5 Consumer Products 22

3.0 PHYSICAL AND CHEMICAL BEHAVIOUR OF TRITIUM IN THE ATMOSPHERE 23
3.1 Physical Behaviour Of Tritium 23
3.1.1 Radioactive Decay 23
3.1.2 Other Physical Properties 23
3.1.3 Mobility in the Environment 25
3.2 Chemical Transformations of Tritium 30
3.2.1 Tritium Transformation from HT to HTO 30
3.2.2 Soil Oxidation of Tritiated CH4 32
3.3 Tritium Behaviour in Atmospheric Plumes 33
3.3.1 Relevant Forms of Tritium 33
3.3.2 Characteristics of Tritium Releases at Nuclear Facilities 34
3.3.3 Dispersion of Tritium Plumes 34
3.3.3.1 Normal Dispersion Processes 34
3.3.3.2 Effects of Topography 36
3.3.3.3 Plume Rise 37
3.3.3.4 Stack and Building Wakes 38
3.3.3.5 Shoreline Fumigation 40
3.3.4 Dry and Wet Deposition 40
3.3.5 Next Generation Dispersion Models 42
3.4 Impact of Climate Change on Tritium Dispersion 42

4.0 DYNAMIC BEHAVIOUR OF TRITIUM IN THE HYDROLOGICAL CYCLE 45
4.1 Partitioning from Air to Soil and Surface Water 45
4.1.1 Partitioning from Air to Soil Water 46
4.1.2 Partitioning from Air to Surface Water 49
4.2 Tritium Transport from Soil Water to Ground Water 51
4.3 Tritium Behaviour in Lake and River Plumes 54

5.0 PREDICTED AND OBSERVED ENVIRONMENTAL BEHAVIOUR OF TRITIUM AT RELEASE SITES 57
5.1 Tritium Light Manufactureres 57
5.1.1 SRB Technologies Limited 57
5.1.2 Shield Source Incorporated 69
5.2 Nuclear Power Stations 73
5.2.1 Pickering (PN) 73
5.2.2. Darlington (DN) 77
5.2.3 Bruce (BN) 80
5.3 Review of Model Performance 83
5.3.1 Air 83
5.3.2 Soil Water 84
5.3.3 Pond Water 84
5.3.4 Groundwater 84

6.0 CONCLUSIONS AND RECOMMENDATIONS 85
6.1 General Conclusions 85
6.2 Recommendations 85

7.0 REFERENCES 86

LIST OF TABLES

2.1 Natural Tritium Distribution (from Begemann, 1963) 5
2.2 Estimated Rates of Generation of Tritium and of its Release in Effluent Streams for Different Types of Reactors 13
2.3 Tritium Releases per MW(e)a for Canadian HWR Power Plants 18
3.1 Atomic Weights of Different Hydrogen Isotopes Contributing to Isotopic Fractionation in the Environment 24
3.2 Thermodynamic Properties of the Oxides of Hydrogen Isotopes 24
3.3 Regional Default Values for P11a, Transfer from HT to HTO in Air (Unitless) 31
3.4 Absolute Humidity, Ha (L • m-3) Averaged over Various Seasons of the Year at Stations Close to Canadian (CANDU) Facilities 32
3.5 Stack and Building Parameters Relevant to Atmospheric Dispersion of Tritium from Canadian Reactor Sites 39
4.1 Values of RFsw Measured at Pickering NGS 47
4.2 Values of RFsw Measured at Three Reactor Sites (IAEA, 2003) 47
4.3 Regional Absolute Humidity and Tritium Transfer from Air to Soil Porewater 48
4.4 Precipitation and Air Monitoring Data Relevant to Air to Pond Transfer Factor 50
4.5 Simple Model of Aquatic Dilution with Distance (Local Plume) Superimposed on Circulating Background for Lake Ontario 56
5.1 Modelled and Measured Tritium in Air at SRBT 60
5.2 Modelled and Measured Tritium in Soil Water at SRBT 61
5.3 Tritium in Roof Runoff, Stack Drippings and Standing Water near Stacks During Precipitation Events at SRBT 62
5.4 Modelled Tritium in Soil Water at Well Locations at SRBT in Previous Years 62
5.5 Modelled and Measured Tritium in Groundwater in SRBT Monitoring Wells 63
5.6 Modelled and Measured Tritium in Air at SSI in 2007 70
5.7 Modelled and Measured Tritium in Ponds at SSI in 2007 70
5.8 Modelled and Measured Tritium in Air at PN, 2007 75
5.9 Modelled Tritium in Soil Water Compared to Measured Tritium in Rain Water at PN, 2007 75
5.10 Modelled and Measured Tritium in Air at DN, 2007 78
5.11 Modelled Tritium in Soil Water Compared to Measured Tritium in Rain Water at DN, 2007 79
5.12 Modelled and Measured Tritium in Air at BN, 2006 81
5.13 Modelled Tritium in Soil Water Compared to Measured Tritium in Rain Water at BN, 2006 82

LIST OF FIGURES

2.1 Origin and Distribution of Tritium in the Hydrological Cycle 6
2.2 Properties of the Standard Atmosphere Depicting the Variation in the Height of the Tropopause with Latitude 10
2.3 Main Nuclear Reactions that Generate Tritium in Reactors 12
2.4 Tritium Production Processes 15
2.5 Depiction of the Process of Ternary Fission of 235U in Reactor Fuel 16
3.1 Conceptual Model Depicting the Hydrological Cycle 27
3.2 Illustration of Atmospheric Plume Dispersion Processes 37
3.3 Environmental Tritium Processes 41
3.4 Predicted Average Winter and Summer Precipitation for the Years 2080-2099 43
4.1 Environmental Transfer Model 46
4.2 Schematic of a Groundwater Flow System 52
4.3 Changes over Time in the Depth Profile of Tritium in Groundwater 54
5.1 Air Sampling Locations at SRBT 64
5.2 Modelled vs Measured Tritium in Air at SRBT, 2006 65
5.3 Soil Water and Groundwater Sampling Locations at SRBT 66
5.4 Tritium Concentrations in Soil Water and Groundwater at MW07-21 on the SRBT Site 67
5.5 Modelled vs Measured Tritium in SRBT Wells, 2007 68
5.6 Modelled vs Measured Tritium in Air at SSI – 2007 Data 71
5.7 Ratio of Predicted to Measured Air vs Distance at SSI – 2007 Data 72
5.8 Modelled vs Measured Tritium in Ponds at SSI in 2007 73
5.9 Modelled vs Measured Tritium in Air at PN, 2007 76
5.10 Modelled Tritium in Soil Water vs Measured Tritium in Rain Water at PN, 2007 .. 77
5.11 Modelled vs Measured Tritium in Air at DN, 2007 79
5.12 Modelled Tritium in Soil Water vs Measured Tritium in Rain Water at DN, 2007 .. 80
5.13 Modelled vs Measured Tritium in Air at BN, 2006 82
5.14 Modelled Tritium in Soil Water vs Measured Tritium in Rain Water at BN, 2007 .. 83

EXECUTIVE SUMMARY

In January 2007, the Canadian Nuclear Safety Commission (CNSC) Tribunal directed CNSC staff to initiate research studies on tritium releases in Canada. In response, staff initiated a "Tritium Studies" project with several information gathering and research activities extending to 2010. The objective of this major research project is to enhance the information used in the regulatory oversight of tritium processing and tritium releases in Canada. The Investigation of the Environmental Fate of Tritium in the Atmosphere *report is part of this project. This review has been prepared for the CNSC by EcoMetrix Incorporated, in association with RWDI Air Inc.*

This report reviews the literature on environmental fate and behaviour of tritium in the atmosphere, including tritium transfer to and behaviour in the hydrological environment. It begins with a description of the various anthropogenic and natural sources of tritium in the atmosphere, the chemical forms of tritium in the atmosphere, and the physical and chemical behaviour of tritium in the atmosphere. The report also describes the dynamic behaviour of tritium in the hydrological cycle, including transfer of tritium from air to soil water and surface water, tritium transport from soil water to groundwater, and tritium behaviour in lake and river receiving environments.

Modelling approaches are described for representing atmospheric dispersion of tritium from point sources, and partitioning of tritium (HTO) from air to soil water, surface water, and groundwater. Modelling was completed at a number of licensed facilities releasing tritium, and model predictions were compared to measured environmental concentrations. Based on these comparisons, our ability to predict environmental concentrations based on current understanding of tritium behaviour is discussed, and factors contributing to model uncertainty are identified.

The sector-averaged Gaussian dispersion model as described in the Canadian Standards Association (CSA) N288.1-08 standard was applied at a number of nuclear power stations and tritium light manufacturing facilities. In all cases, the model was conservative, tending to over-predict the tritium concentration in air. In three cases, predictions were slightly higher than annual average measured values (20-83% higher on average, generally within a factor of 2). In two cases, tritium concentrations were over-predicted more substantially by 2 or 3 times on average. Measured air values are uncertain due to unresolved differences between active and passive air samplers.

Model-predicted tritium in soil water was compared to either measured soil water or measured rain water (since soil water derives from rain water and should be similar). In three cases, predictions were slightly higher than measured values (35-62% on average, generally within a factor of 2). In one case, tritium in soil water was over-predicted more substantially by 2 times on average). It was noted that, when air concentrations are changing rapidly, soil water can lag behind.

Model-predicted tritium in groundwater was compared to measured groundwater at one facility and was 52% higher on average (generally within a factor of 3). It was noted that groundwater wells may be influenced by nearby snow storage, or by horizontal groundwater flow, as well as vertical infiltration, and are subject to local variation in sub-surface conditions. Groundwater lags behind soil water based on well depth and vertical travel time.

Model-predicted tritium in pond water was compared to measured pond water at one facility and was 28% higher on average (generally within a factor of 2). It was noted that ponds and marshes may be subject to up-gradient inflows of soil water or groundwater, and thus may not be at equilibrium with current local air concentrations.

Recommendations include studies to resolve the discrepancies that are often seen between active and passive air sampler results, as well as near-field studies of air, soil water and groundwater designed to better understand the time lags in soil water and groundwater, and the importance of up-gradient effects.

1.0 INTRODUCTION

Elevated concentrations of tritium in the environment associated with producing, handling and managing the radioactive form of hydrogen at nuclear facilities are of great public concern. Under the Nuclear Safety and Control Act (NSCA) the Canadian Nuclear Safety Commission's (CNSC) mandate includes the dissemination of scientific, technical and regulatory information concerning the activities of the CNSC, and the effects on the environment, on the health and safety of persons, of the development, production, possession, transport and use of nuclear substances.

In January, 2007, the Commission tribunal directed CNSC staff to initiate research studies on tritium releases in Canada. In response, CNSC staff initiated a "*Tritium Studies*" project with several information gathering and research activities extending to 2010 (fact sheet available at www.nuclearsafety.gc.ca). The objective of this major research project is to enhance the information available to guide regulatory oversight of tritium processing and tritium releases in Canada. A review of the information available on the environmental fate of tritium in the atmosphere, in the context of controlled releases from licensed facilities, represents activity 5.5 in the *Tritium Studies* project.

The CNSC requested EcoMetrix Incorporated (EcoMetrix) in association with RWDI Air Inc. (RWDI) to prepare this review of the environmental fate of tritium in the atmosphere. The report describes the various anthropogenic and natural sources of tritium in the atmosphere, the chemical forms of tritium in the atmosphere, and the physical and chemical behaviour of tritium in the atmosphere. This includes discussion of atmospheric dispersion processes and modeling approaches to estimating dispersion in the context of public dose assessment. The report also describes the dynamic behaviour of tritium in the hydrological cycle, including transfer of tritium from air to soil water and surface water, tritium transport from soil water to groundwater, and tritium behaviour in lake and river receiving environments.

Beyond this literature review of tritium behaviour in atmospheric and hydrological environments, this report considers available tritium monitoring data for a number of licensed nuclear facilities releasing tritium, examines model predictions of environmental concentrations of tritium, and compares predicted and observed concentrations. Based on these comparisons, our ability to predict environmental concentrations based on current understanding of tritium behaviour is discussed, and factors contributing to model uncertainty are identified.

Following this introduction the report is organized as follows. Section 2.0 describes the various sources of tritium, Section 3.0 describes the physical and chemical behaviour of tritium in the atmosphere, and Section 4.0 describes the behaviour of tritium in the hydrological cycle, and Section 5.0 compares predicted and observed behaviour at various release sites. References cited are included in Section 6.0.

2.0 SOURCES AND CHEMICAL FORMS OF TRITIUM

This section outlines the sources and chemical forms of tritium in the atmosphere. Both natural and anthropogenic sources of tritium are described.

2.1 Chemical Forms

Tritium is a rare but natural isotope of hydrogen (H), and is the only natural hydrogen isotope that is radioactive. Whereas the common hydrogen nucleus contains a single proton, the nucleus of a tritium atom also contains two neutrons (ANL, 2005). Thus, the mass of a tritium atom is 3 times that of an ordinary hydrogen atom. The tritium atom is sometimes designated T to distinguish it from the common lighter isotope.

Notwithstanding the difference in mass, tritium can be found in the same chemical forms as hydrogen. The most important forms, from the perspective of atmospheric behaviour of tritium, are tritiated hydrogen gas (HT) and tritiated water (HTO). These tritiated forms behave chemically like hydrogen gas (H_2) and water (H_2O). When tritium is incorporated into hydrocarbon molecules within an organism, it is referred to as organically bound tritium (OBT). Since OBT is not involved in atmospheric processes, it is not considered further in this review. Tritiated methane (CH_3T) accounts for a small amount of atmospheric tritium.

2.2 Natural Abundance

Hydrogen is the most abundant element in the universe, comprising approximately 90% of the luminous universe by weight. Ordinary hydrogen (^1H) accounts for greater than 99.985% of all naturally-occurring hydrogen, whereas deuterium (^2H) comprises approximately 0.015%. By comparison, tritium (^3H) represents only approximately 10^{-16} percent of hydrogen naturally occurring (Gross *et al.*, 1951).

Natural atmospheric hydrogen has been estimated to contain approximately 4×10^{-15} tritium atoms per hydrogen atom, whereas hydrogen in natural surface waters contains approximately 10^{-18} tritium atoms per hydrogen atom, according to early surveys (Bibron, 1963; Faltings and Harteck, 1950; Fireman and Rowland, 1961; Grosse *et al.*, 1951,1954; Harteck, 1954; Harteck and Faltings, 1950). On this basis, the 'tritium unit' (TU) was defined as 1 tritium atom per 10^{18} atoms of hydrogen (Gross *et al.*, 1951). Today, tritium levels are more commonly reported in activity units of Bq per litre of water, where 1 TU is equivalent to 0.118 Bq/L (e.g., ICRU, 1963).

Tritium levels present in the atmosphere (T/H ratio) are approximately 10^3 to 10^4 times higher than in precipitated rainwater, although hydrogen gas quantities in the atmosphere are 10^4-fold lower than the mean quantity of water vapour (Begemann, 1962; Bibron, 1963; Grosse *et al.*, 1954; Harteck, 1954; Harteck and Faltings, 1950). Global levels correspond to approximately 0.12% of natural tritium in the form of hydrogen gas, approximately 0.1% as water vapour and the remaining 99.78% in the hydrosphere, predominantly in the oceans (Begemann, 1962; Jacobs, 1968). Of the total natural tritium inventory, almost 99% has been estimated to occur in the oceans, with less than 1% occurring in the atmosphere and in the

groundwater (Table 2.1). Under undisturbed natural conditions the tritium concentration in precipitation was likely approximately 5 TU, which is equivalent to a specific activity of about 0.6 Bq/L (Roether, 1967). By comparison, tritium is naturally present in surface waters at concentrations of approximately 0.37 to 1.11 Bq/L (ANL, 2005).

2.3 Natural Sources

Tritium is generated by both natural and artificial processes (Figure 2.1). Tritium is naturally produced primarily through the interaction of cosmic radiation protons and neutrons with gases (including nitrogen, oxygen and argon) in the upper atmosphere (Casaletto *et al.*, 1962; Dorfman and Hemmer, 1954; Eisenbud and Gesell, 1997; Suess, 1958; Thompson and Schaeffer, 1955; UN, ILO and WHO, 1983; Yang and Gevantman, 1960, 1964; Zerriffi, 1996). Approximately two-thirds of the natural tritium is produced in the stratosphere, with much lower rates of production in the hydrosphere and in the lithosphere at the surface of the earth (UN, ILO and WHO, 1983). In addition, some tritium may be generated in the extra-terrestrial environment and enter the atmosphere along with cosmic rays or solar wind (UN, ILO and WHO, 1983). Most of the natural tritium is found in the environment as tritiated water (generally designated as HTO) is in the liquid or vapour state.

Table 2.1: Natural Tritium Distribution (from Begemann, 1963).

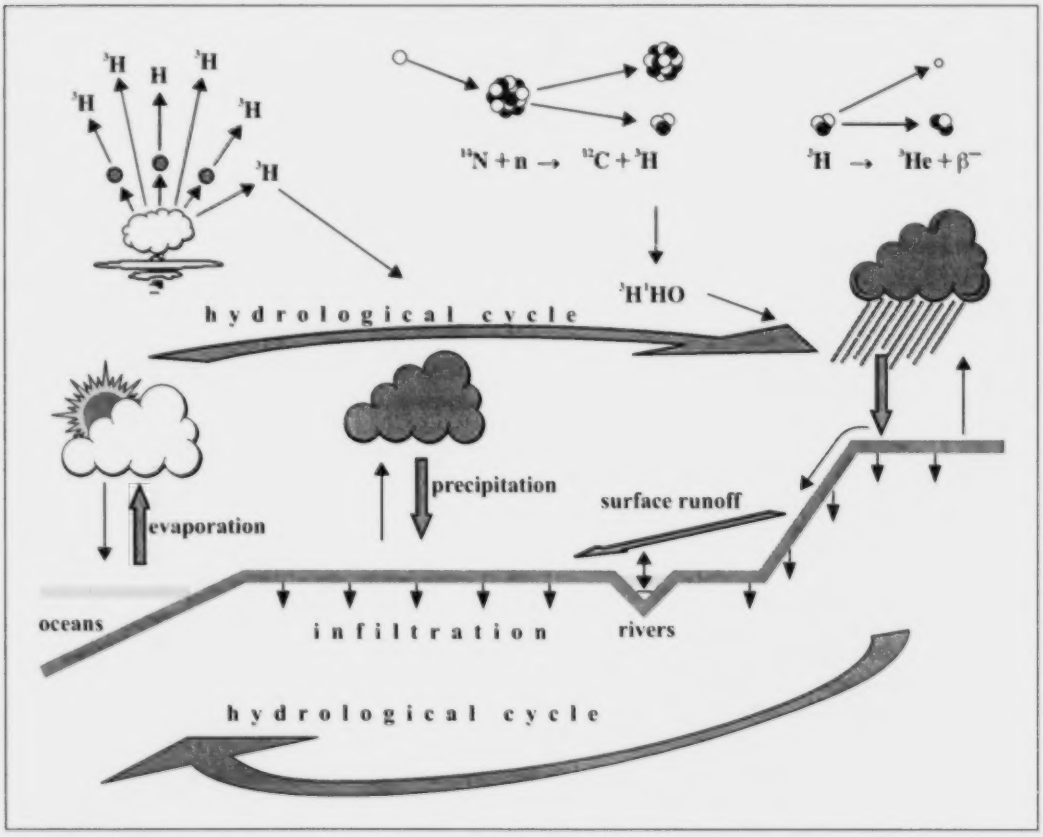
Environmental Compartment	Percent Tritium Distribution (%)
Hydrosphere	~90
Troposphere	
Water vapour	0.1
Molecular hydrogen	0.02 to 0.2
Methane	< 0.04
Stratosphere	
Water	~10
Molecular hydrogen	0.004 to 0.007

The most important of the atmospheric reactions involves the interaction between a fast neutron (> 4 MeV) and atmospheric nitrogen (Young and Foster, 1972), as described by:

$$^{14}_{7}N + {}^{1}_{0}n \rightarrow {}^{12}_{6}C + {}^{3}_{1}T \qquad \text{Equation 2.1}$$

Natural tritium production per unit time and per unit area at the earth's surface is estimated to occur at a rate of approximately 0.1 to 1.3 tritium atoms/cm^2/sec, with the most probable values occurring around 0.5 to 1.0 tritium atoms/cm2/sec (Begemann, 1958, 1962; Begemann and Libby, 1957; Bolin, 1958; Craig, 1957; Craig and Lal, 1961; Currie, 1959; Currie *et al.*, 1956; Flamm *et al.*, 1962; Hagemann *et al.*, 1959; Jacobs, 1968; Libby, 1952; Newkirk, 1963; UN, 1960; UNSCEAR, 1977; von Buttlar, 1963; von Buttlar and Libby, 1955; Wilson and Ferguson, 1960; Zahn *et al.*, 1998).

Figure 2.1: Origin and Distribution of Tritium in the Hydrological Cycle. The turnover of tritium is very fast, except where it is fixed in glacier ice or groundwater, and when it has been incorporated into organically-bound tritium (OBT) in organisms (from http://www.iaea.org/programmes/ripc/ih/volumes/vol_one/cht_i_08.pdf).



A small fraction of tritium may also originate in the stratosphere from extra-terrestrial sources, such as direct ejection by solar flares and by stars, although fast-neutron irradiation would be the dominant source, producing approximately 10-fold more tritium than the former (Fireman, 1953; Flamm *et al.*, 1962; Lal and Peters, 1967). For example, solar flares have been estimated to generate an average of 0.1 tritium atom/cm^{2}/sec at the Earth's surface over the course of the solar cycle (Flamm *et al.*, 1962).

Trace amounts of tritium can also be formed through slow-neutron reactions on ^{6}Li in the oceans and the lithosphere, neutron irradiation of deuterium and reactions of neutrons produced during spontaneous fission in terrestrial material (Jacobs, 1968; UNSCEAR, 1977). Such processes have been estimated to result in tritium production rates of 10^{-3} atoms/cm^2/s in the lithosphere and at 10^{-6} tritium atoms/cm^2/sec in the hydrosphere (Fireman, 1953; Kaufman and Libby, 1954; UN, ILO and WHO, 1983). It is expected that ecosystem changes generated from processes such as climate change may influence natural rates of tritium production and its fate in the atmosphere, hydrosphere and lithosphere.

Tritium production through natural processes has resulted in a global steady-state inventory of approximately 2.65 kg, corresponding to about 9.6 x 10^5 TBq (NCRP, 1979; Zerriffi, 1996). Due to the relatively short half-life of tritium, of 12.32 years, tritium produced in this manner does not accumulate over geological time-scales, which explains its negligible natural abundance (Lucas and Unterweger, 2000).

2.4 Anthropogenic Sources

In addition to its natural sources, tritium also has a number of anthropogenic sources which account for the dominant proportion of the global tritium inventory. Anthropogenic tritium sources include fallout from nuclear weapons testing, nuclear reactors, future fusion reactors, fuel reprocessing plants, heavy water production facilities and commercial production for medical diagnostics, radiopharmaceuticals, luminous paints, sign illumination, self-luminous aircraft, airport runway lights, luminous dials, gauges and wrist watches, and others (Galeriu *et al.*, 2005; Weiss *et al.*, 1979a; UN, ILO and WHO, 1983). Commercial uses of tritium account for only a small fraction of the tritium used worldwide. Instead, the primary use of tritium has been to boost the yield of both fission and thermonuclear (or fusion) weapons, increasing the efficiency with which the nuclear explosive materials are used (Kalinowski and Colschen, 1995).

2.4.1 Thermonuclear Detonation during Nuclear Weapons Testing

Nuclear tests have been conducted in the atmosphere since 1945, producing tritium in amounts that greatly exceed the global natural activity, particularly during 1954 to 1958 and 1961 to 1962 when a number of large-yield test series were undertaken (UN, ILO and WHO, 1983). The tritium activity arising from atmospheric nuclear tests can be estimated from the fission and fusion yields of the weapons tests or from environmental measurements. For example, the tritium activity produced per unit yield is dependent upon the attributes of the device, as well as on the characteristics of the detonation site, and tritium generation from fusion reactions is much higher than from fission (NCRP, 1979).

Tritium is produced as a fission product in nuclear weapons tests and in nuclear power reactors, with a yield of approximately 0.01% (Albenesius, 1959; Albenesius and Ondrejcin, 1960; ANL, 2005; Haney *et al.*, 1962; Horrocks, 1964; Jacobs, 1968; Serot *et al.*, 2005; Sloth *et al.*, 1962; Watson, 1961; Wegner, 1961). This represents approximately one tritium atom per 10,000 fission events. A residual of approximately 0.7 to 5.0 kg of tritium per megaton-equivalent explosion is generated from the fusion reaction of a deuterium-tritium bomb in a pure thermonuclear detonation, with an additional yield of up to 0.15 kg of tritium through the neutron irradiation of nitrogen (as shown in Equation 2.1 above) (Eriksson, 1965; Leipunsky, 1960; Martell, 1959, 1963; Miskel, 1964).

Total and fission tritium yields for each reported atmospheric test over the period between 1945 and 1978 have been estimated (Bennett, 1978). Data compiled over this period included tritium releases from 422 nuclear tests in the atmosphere, with cumulative yields of 217 megatons and 328 megatons for fission and fusion, respectively. This corresponded to estimated yields of 2.6×10^{13} Bq tritium per megaton for fission explosions and 7.4×10^{17} Bq tritium per megaton for fusion, and a total tritium activity of 2.4×10^{20} Bq tritium for atmospheric tests (based on tritium generated from fusion reactions) (Miskel, 1973).

The tritium that is produced by a nuclear explosion is almost completely converted to tritiated water (HTO), which then mixes with environmental water, as described by the following equation (UN, ILO and WHO, 1983):

$$HT + H_2O \rightleftharpoons HTO + H_2 \qquad \text{Equation 2.2}$$

Most of the bomb tritium, which has been oxidized to form water, is removed from the troposphere by precipitation, although the tritium-to-hydrogen ratio for atmospheric hydrogen gas and methane is much higher (Jacobs, 1968; Martell, 1959). As a result, past thermonuclear detonations have led to sharp increases in tritium levels of up to several orders of magnitude greater than natural levels in rain, with temporary increases in tritium of 1000-fold in precipitation in the northern hemisphere following weapons testing in the early 1960s (e.g., Begemann and Libby, 1957; von Buttlar and Libby, 1955). Since 1963, the temporary extremes in the tritium content have decreased to essentially natural values in the winter and values that are approximately twice natural levels in the summer.

Based on measurements of tritium in precipitation taken at stations in the IAEA network, a total tritium production of 1.7×10^{20} Bq from weapons testing has been estimated (Schell *et al.*, 1974). Similar weapons tritium estimates, ranging from 1.2×10^{20} Bq (in oceans only) to 2.4×10^{20} Bq, have been made using measurements of tritium taken in vertical profiles in the oceans (Bowen and Roether, 1973; Mitchel, 1976; Ostlund and Fine, 1979; UNSCEAR, 1977). On the basis of these estimates, UNSCEAR (1977) adopted a value of 1.7×10^{20} Bq tritium to represent the amount of tritium that was dispersed globally due to atmospheric weapons testing up to 1976.

When tritium is injected into the stratosphere, scavenging by precipitation is not as quick as in the troposphere (Stebbins, 1961). By comparison, underground fusion explosions produce tritium primarily in the form of tritiated water that then moves with the groundwater (Stead, 1963).

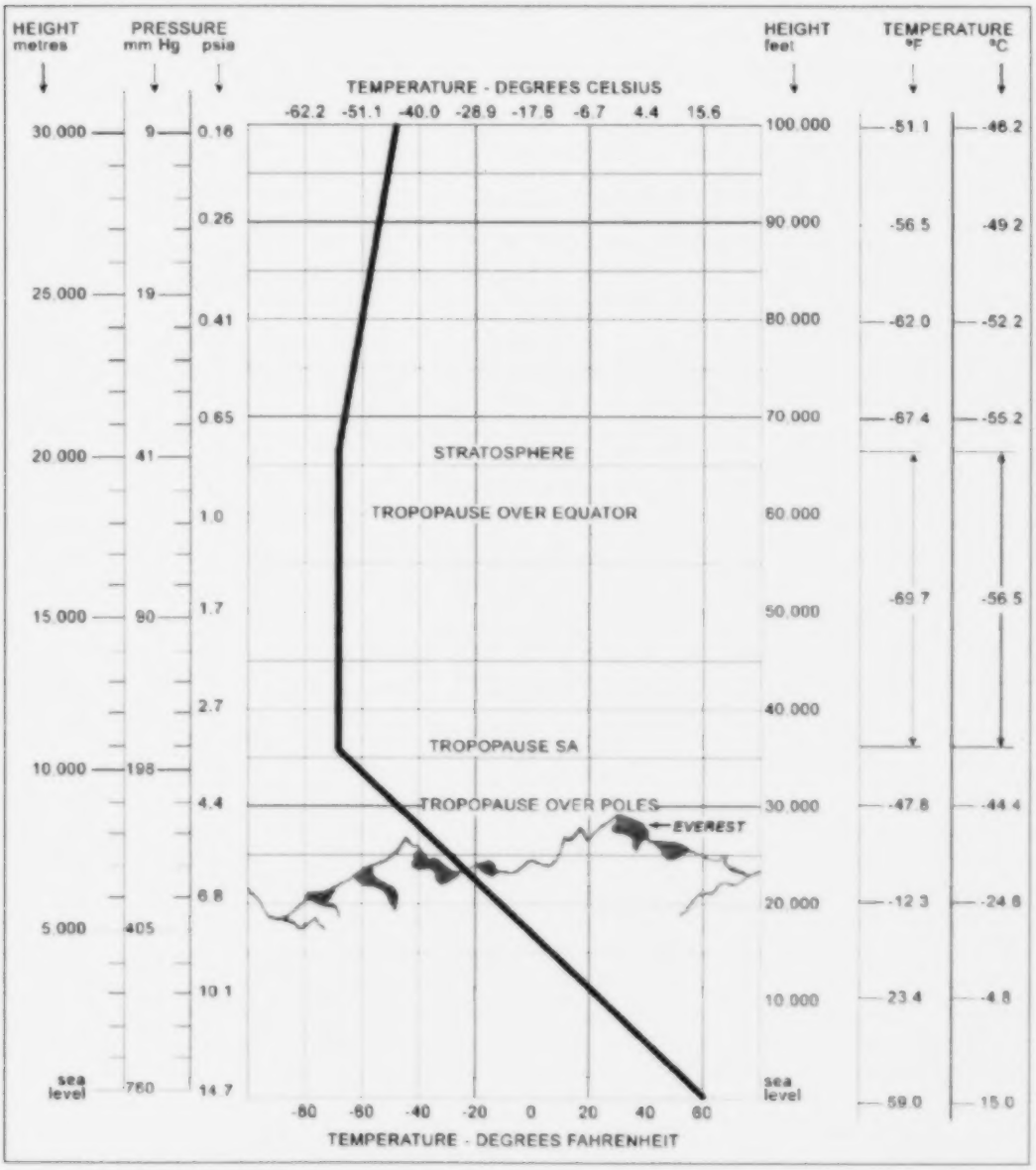
In general, partitioning of radioactive debris between the stratosphere and the troposphere varies with the height of the shot, the latitude, and the yield (Martell, 1963). For example, high-altitude thermonuclear explosions in tropical regions will introduce tritium directly into the stratosphere where mixing processes are very slow (Bibron, 1965; Jacobs, 1968; Libby, 1958). By comparison, low-altitude detonations in tropical regions result in the entrainment of a substantial fraction of the water vapour in the environmental atmosphere by the ascending air current that is created by the explosion. The tropopause then serves as a cold trap, preventing most of the water vapour from penetrating into the stratosphere. The ice particles that are formed at the tropopause then fall back rapidly into the troposphere.

In the polar regions, the natural water vapour content of the air is relatively low, and therefore, much of the tritium from detonations in polar regions will penetrate into the stratosphere, later moving to lower latitudes before moving into the troposphere. Therefore, it is expected that any changes in the water vapour content, air temperature (and corresponding capacity of the air mass to hold water), global cycling of the air masses, as well as any changes impacting the hydrological cycle, in general, would be expected to affect the fate of tritium in the environment. For example, climate change (which is discussed in more detail in Section 1.5 below) has the potential to measurably influence tritium cycling both locally and globally.

The distribution of tritium that is produced by nuclear detonations occurs in roughly the same manner as for natural tritium (Libby, 1963). HTO is the predominant form of the tritium, since it is rapidly produced under the oxidizing conditions of the thermonuclear fireball (Bibron, 1963; Bishop *et al.*, 1962). In addition, the reducing medium at the centre of the fireball results in the production of highly tritiated hydrogen and methane (Wolfgang, 1961).

Approximately 20% of the radionuclide's released to the atmosphere during a ground surface detonation are injected into the stratosphere, whereas approximately 80% of those released at a water surface end up reaching the stratosphere (Bibron, 1965). Stratospheric tritium is slowly released into the troposphere (with a mean residence time of slightly over 1 year to 10 years in the stratosphere), after which tritium removal as water vapour occurs much more rapidly through the flushing action of rainfall (Barrett and Huebner, 1961; Begemann, 1962; Eriksson, 1965; Gat *et al.*, 1962; Hagemann *et al.*, 1959; Jacobs, 1968; Libby, 1958; Martell, 1963; Stebbins, 1961).

Figure 2.2: The Properties of the Standard Atmosphere Depicting the Variation in the Height of the Tropopause with Latitude (from Transport Canada, 2004).



Transport of air masses and debris from the stratosphere to the troposphere arises from the action of turbulent diffusion caused by currents of horizontal circulation that lead to seasonal changes in the tritium content of rain (Bibron, 1965). Extremely fast jet currents, particularly in sub-tropical reaches, can initiate relatively important exchanges that facilitate quite rapid north-to-south mixing in the stratosphere (Libby, 1958). Therefore, tritium becomes more homogeneously mixed in this way (Begemann, 1958). These jet currents reach their maximum intensity at the end of the winter and in the spring, leading to seasonal variations in tritium fallout, with maximum values occurring in the mid-latitudinal areas during the spring and early summer as tritium is washed from the stratosphere into the troposphere (Libby, 1963; Taylor, 1964; Taylor *et al.*, 1963). Specifically, it has been estimated that approximately half of the tritium originating from the detonation of thermonuclear devices falls in the zone between 30° and 50° latitude (Libby, 1963).

Following entry into the troposphere, vertical mixing becomes quite pronounced, leading to a rapid flushing out by rains corresponding to a relatively short tritium residence time in the troposphere (approximately 21 to 40 days) compared to the one to 10 year residence time in the stratosphere (Barrett and Huebner, 1960, 1961; Begemann and Libby, 1957; Bolin, 1964; Brown and Grummitt, 1956; Libby, 1958, 1963; Walton *et al.*, 1962). Tritium transport can also occur through eddy diffusion, the rate of which is dependent upon air motion, the rate of evaporation over water bodies, as well as the relative humidity of the air (Engelke and Hemis, 1962; Eriksson, 1965).

2.4.2 Nuclear Reactors

Tritium is produced by a number of processes in reactors, as depicted in Figure 2.3 below and as discussed in the sections that follow. In general, these reactions can be sub-divided into tritium production by ternary fission (or fission of the reactor fuel); and neutron activation reactions with lithium and boron isotopes dissolved in or in contact with the primary coolant, or with naturally-occurring deuterium in the primary coolant (Estournel, 1962; UN, ILO and WHO, 1983).

The relative importance of these reactions in the production of tritium is dependent upon the type of reactor and its design. For the purposes of this report, four reactor designs were discussed from the perspective of tritium generation. These included pressurized-water reactors (PWRs), boiling water reactors (PWRs), heavy water reactors (HWRs) and gas-cooled reactors (GCRs). The relative tritium contributions of each design with respect to tritium generation and tritium release via effluent streams have been summarized in Table 2.2.

Additional information with respect to the potential for tritium generation and release will be available in the near future for thermonuclear (or fusion) reactors, which are currently under development (e.g., ITER; http://www.iter.org/).

Figure 2.3: Main Nuclear Reactions that Generate Tritium in Reactors (from UN, ILO and WHO, 1983).

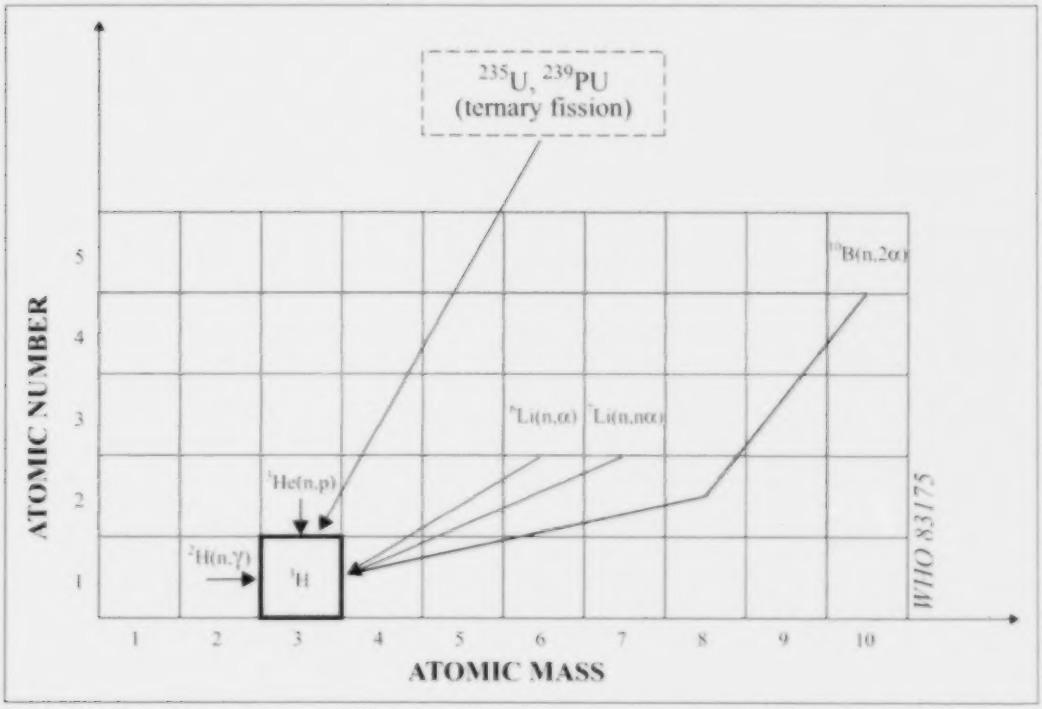


Table 2.2: Estimated Rates of Generation of Tritium and of its Release in Effluent Streams for Different Types of Reactors (10^{10} Bq per MW(e)a) (Gratwohl, 1973; Kouts and Long, 1973; Smith and Gilbert, 1973; Trevorrow *et al.*, 1974; UNSCEAR, 1977).

	Rate of Tritium Generation and Release by [a] Reactor Type (10^{10} Bq per MW(e)a)							
	PWR		BWR		HWR		GCR	
Source	Generation	Effluent Stream	Generation	Effluent Stream	Generation	Effluent Stream	Generation	Effluent Stream
Fission	75	< 0.7	75	< 0.7	55	< 0.6	75	< 0.7
Activation								
Deuterium	0.004	0.004	0.04	0.04	2000	[b] 75		
Lithium	0.07	0.07					2	0.4
Boron	2.6	2.6		30	0			
Rounded Total	**80**	**3**	**110**	**0.5**	**2000**	**75**	**80**	**1**

[a] *PWR: Pressurized water reactor; BWR: Boiling water reactor; HWR: Heavy water reactor; GCR: Gas-cooled reactor*

[b] *Depending on the irradiation time and on the net leakage of heavy water. This release per MW(e)a is typical for HWR type reactors over the 1990-94 period, according to UNSCEAR (2000), Annex C, Fig. XVI. There is approximately order of magnitude variability around this value.*

2.4.2.1 Pressurized Water Reactors (PWRs)

In pressurized water reactors (PWRs), tritium is produced primarily through neutron capture by boron-10 (^{10}B) in the primary coolant (water), resulting in the production of 2.6 x 10^{10} Bq tritium per MW(e)a (UN, ILO and WHO, 1983). Boric acid or 'soluble boron' is added to the PWR reactor coolant system (RCS) as a soluble reactivity shim, which in effect, reduces the rate at which fission is occurring in the reactor core by absorbing neutrons to control the reactivity (Jacobs, 1968).

Neutron capture by ^{10}B produces a number of outcomes, as described by the following equations (Jones, 2007):

$$^{10}_{5}B + {}^{10}_{5}B \rightarrow [{}^{11}_{5}B]^{*} \rightarrow {}^{7}_{3}Li + {}^{4}_{2}He \qquad \text{Equation 2.3}$$

$$^{10}_{5}B + {}^{1}_{0}n \rightarrow [{}^{11}_{5}B]^{*} \rightarrow {}^{3}_{1}H + 2\cdot({}^{4}_{2}He) \qquad \text{Equation 2.4}$$

Of these processes, the first (as shown in Equation 2.3) describes the production of lithium in the form of lithium hydroxide (LiOH), which is used in PWR reactor coolant pH control (Locante and Malinowski, 1973). The maintenance of 2 parts per million (ppm) lithium hydroxide in the reactor coolant results in the formation of approximately 7×10^8 Bq tritium per MW(e)a (UN, ILO and WHO, 1983). By comparison, Equation 2.4 presents tritium production from ^{10}B through neutron capture.

Neutron capture by ^7Li (which is generated via the process shown in Equation 2.3 above), is also a minor source of tritium (Jones, 2007), as follows:

$$^7_3Li + {}^1_0n \rightarrow {}^3_1H + {}^4_2He + {}^1_0n \qquad \text{Equation 2.5}$$

In addition, lithium-based cation exchangers can be used for cooling-water purification, which may result in quite high tritium production (Jacobs, 1968; IAEA/WHO, 2004a, 2004b). For thermal and slow neutrons, the production of tritium from lithium is due to the ^6Li isotope (as depicted in Figure 2.4 below). For fast neutrons, ^7Li plays the principal role, as per the reaction shown in Equation 2.5 above (Verzaux, 1952). A changeover from natural LiOH resin to ^7LiOH resin can reduce the tritium concentration in the primary coolant by two orders of magnitude (Lechnick, 1962).

Ninety percent of the total tritium in PWR reactor coolant is produced in the coolant by the soluble boric acid reactivity shim. The remaining 10% is produced by other processes, including ternary fission, ^{10}B burnable poisons, ^6Li neutron capture, and deuterium or ^3He activation following thermal-neutron irradiation in the reactor coolant (Estournel, 1962; Jacobs, 1968; Sültenfuß, 2008). For example, Figure 2.4 depicts two basic nuclear processes that lead to the production of tritium, which include a reactor and an accelerator process.

Boron-10 is also present with boron in PWR fuel assemblies as a burnable poison or neutron absorber to maintain a relative constant reactivity over the life of the reactors and to avoid the use of control rods, which locally depress the neutron flux (US-DOE, 1993). The annual production of fission product tritium in the fuel rods of a pressurized water reactor (PWR) is in the range of 6 to 9×10^{11} Bq per MW(e)a (NCRP, 1979).

In general, environmental tritium discharges from PWRs are dependent upon waste management practices, as well as the materials used in a reactor (UN, ILO and WHO, 1983; IAEA/WHO, 2004a, 2004b). For example, large differences in tritium releases can occur between PWRs due to the type of fuel cladding used. Of the tritium produced in the fuel rods, a small percentage (comprising 1% or less) is expected to be released into the coolant through defects in the cladding, for newer reactor designs, which employ zirconium alloy cladding. For example, total combined releases of approximately 3×10^{10} Bq tritium per MW(e)a for liquid effluents and of 10^9 Bq per MW(e)a for airborne effluents have been reported for nine newer PWRs with zirconium alloy-clad fuel (including contributions from all nine reactors). In contrast, earlier PWR designs, which used stainless steel cladding, tend to release most of the tritium that has been produced in the fuel through the process of diffusion (Jacobs, 1968). For example, one older PWR with stainless steel-clad fuel was

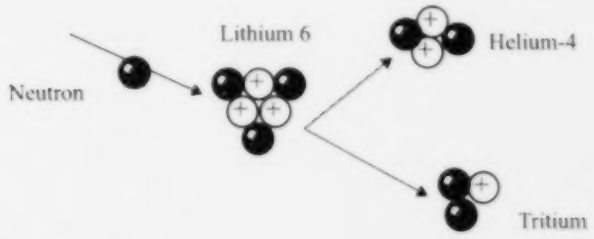
reported to release approximately 4 x 10^{11} Bq tritium per MW(e)a in its liquid effluents and approximately 4 x 10^{10} Bq per MW(e)a in airborne effluents (Kahn *et al.*, 1974), representing values that were higher than the cumulative releases of the nine newer PWR reactors, as described above.

Tritium is also produced by ternary fission of U-235, but only a small fraction (≈1%) of the total H-3 produced in the fuel is believed to diffuse through the cladding into the coolant (Lin, 1996; Figure 2.5).

Figure 2.4: Tritium Production Processes (from Zerriffi, 1996).

Reactor Process:

A neutron strikes a Lithium-6 nucleus making a Tritium nucleus a Helium-4 nucleus.



Accelerator Process:

A neutron strikes a Helium-3 nucleus making a Tritium nucleus and a proton.

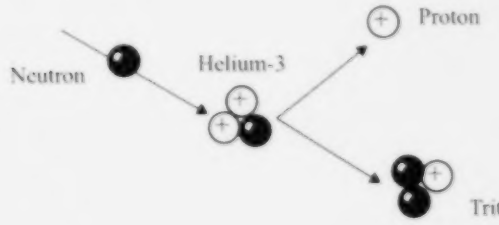
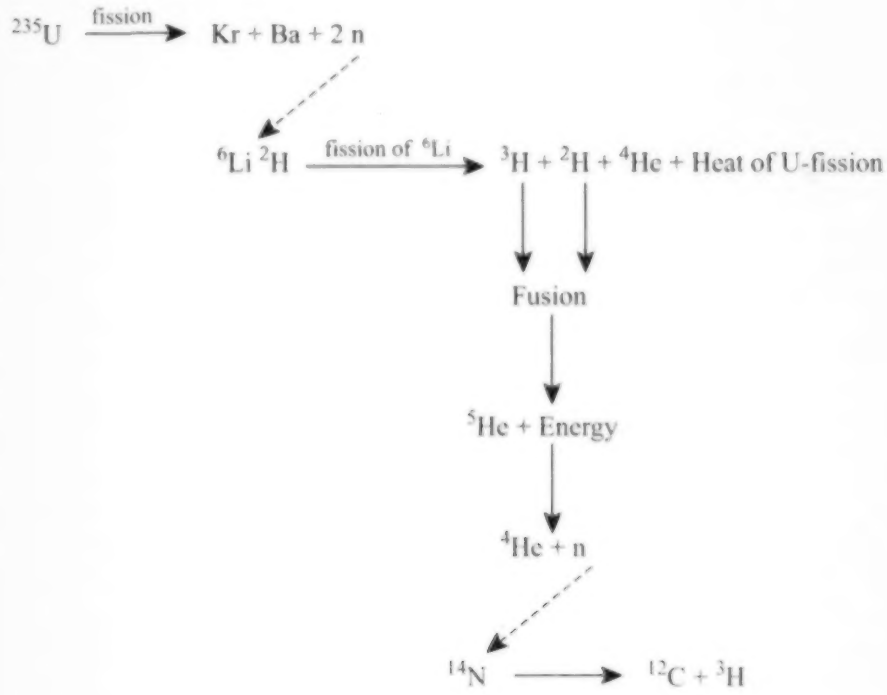


Figure 2.5: Depiction of the Process of Ternary Fission of ^{235}U in Reactor Fuel (from Sültenfuß, 2008).



2.4.2.2 Boiling Water Reactors (BWRs)

Tritium in Boiling Water Reactors (BWRs) is primarily produced from burnable poison, ternary fission, and deuterium activation. Tritium is produced in BWRs by ternary fission in the fuel at approximately the same rate as in PWRs (i.e., at a rate of 6 to 9 x 10^{11} Bq tritium per MW(e)a). In addition, tritium can be generated by neutron activation in both the coolant and in the control rods (UN, ILO and WHO, 1983). For example, tritium is generated in the coolant of BWRs by activation of deuterium at a rate of about 4 x 10^{8} Bq per MW(e)a, as described by the following equation:

$$^{2}_{1}H + ^{1}_{0}n \rightarrow ^{3}_{1}H \qquad \text{Equation 2.6}$$

In terms of tritium production by neutron activation in the control rods, prior to 1971, boron carbide control rods were used in BWRs, yielding approximately 3 x 10^{11} Bq tritium per MW(e)a (Smith and Gilbert, 1973), although tritium has not been shown to diffuse through the boron carbide matrix (Trevorrow *et al.*, 1974). As for PWRs, current BWRs use zirconium alloy cladding, which limits the tritium release into the coolant to less than 7 x 10^{9} Bq per MW(e)a.

Tritium production and the corresponding activity concentrations discharged from BWRs into the environment are lower than those of PWRs, representing approximately one tenth the total tritium generated by a PWR due to the absence of boric acid in the coolant (Jones, 2007). In general, this leads to less tritium being produced in or diffusing into the primary coolant of BWRs, and mean discharge rates of 4×10^9 and 2×10^9 Bq tritium per MW(e)a in liquid and airborne effluents, respectively (UNSCEAR, 1977).

Tritium and other radionuclide emissions are further reduced, since BWRs are designed to reprocess the liquid wastes that are generated, resulting in very little to no liquid effluents, including tritium. Lower tritium production in BWRs compared to PWRs facilitates the recovery of liquid waste without building up tritium to total levels in excess of operating limits.

2.4.2.3 Heavy Water Reactors (HWRs)

Heavy water (D_2O) can be used as a moderator, a reflector and a coolant in heavy water reactors (HWRs) (Butler, 1963; Jackson, 1964; Holmquist, 1965; Patterson, 1958; Matic-Vukmirovic, 1965). Such reactors produce tritium predominantly by neutron bombardment of deuterium (as described in Equation 1.6), with negligible tritium production due to penetration of fission products through thin fuel element claddings and induced tritium activity from the heavy water (Jacobs, 1968; Table 2.2).

The tritium concentration in the heavy water is a function of the reactor power level and the irradiation time, where tritium losses from the heavy water are quite low (Matic-Vukmirovic, 1965). In general, three reactions play a role in the tritium chemistry in HWRs (Jacobs, 1968). These include: 1.) The dissociation of the DTO that is formed by neutron irradiation to form tritium radicals that further react with deuterium radicals to form DT; 2.) The reaction of D_2O with DT to produce DTO and D_2; and 3.) the formation of hydrogen molecules that are depleted in tritium due to the two-fold higher probability of forming deuterium radicals compared to tritium radicals during the radiolysis of DTO (Jacobs, 1968). Of these processes, the first leads to gaseous tritium discharges, whereas the latter two processes tend to favour tritium retention.

Although the amount of tritium generated in fuel of HWRs by ternary fission of the reactor fuel is approximately the same as for light water reactors (such as PWRs and BWRs), it is largely exceeded by the tritium production that occurs in the D_2O coolant and moderator through neutron activation, which has been estimated to be approximately 2×10^{13} Bq per MW(e)a (Kouts and Long, 1973; UN, ILO and WHO, 1983).

External tritium losses from HWRs tend to occur as releases to the atmosphere from the reactor stack, as well as releases to the water via effluent and groundwater, both mainly as HTO (Jacobs, 1968). Environmental discharges are dependent upon the potential for D_2O leakage, as well as the tritium activity in the reactor coolant and moderator, which builds up with the irradiation time (Jacobs, 1968; UN, ILO and WHO, 1983). Such releases contribute to anticipated annual losses of 0.5% to 3%, corresponding to normalized tritium release rates

of approximately 6 x 10^{11} and 1.5 x 10^{11} Bq per MW(e)a for airborne and liquid effluents, respectively, based on estimated releases over the life-time of the reactor (Gratwohl, 1973; UNSCEAR, 1977). In addition, when heavy water serves as both a coolant and a moderator, the complex system of heat exchangers and lines can lead to line breakages and occasional spills, which may introduce tritium to the environment (Jacobs, 1968).

UNSCEAR (2000) presents tritium release data for HWR reactors over the 1990-94 period, which supports a typical total release of 7.5 x 10^{11} Bq/MW(e) a. There is approximately order of magnitude variability around this value. Table 2.3 presents more recent tritium release data for Canadian CANDU generating stations. Most of these stations are releasing less than 7.5 x 10^{11} Bq/MW(e) a, but still within the range cited by UNSCEAR (2000).

Table 2.3: Tritium Releases per MW(e)a for Canadian HWR Power Plants

Generation or Release	Pickering	Darlington	Bruce	Gentilly-2	Pt. Lepreau
Power Generation (MW(e)a)	3094	3542	6516	675	640
Year of Tritium Release:	2006 [1]	2006 [1]	2004 [2]	2002 [3]	2002 [3]
Release to Air (Bq/a)	5.7 x 10^{14}	2.25 x 10^{14}	8.97 x 10^{14}	1.8 x 10^{14}	1.3 x 10^{14}
Release to Water (Bq/a)	3.3 x 10^{14}	1.9 x 10^{14}	5.84 x 10^{14}	5.0 x 10^{14}	1.4 x 10^{14}
Normalized Release (Bq/MW(e)a)	2.91 x 10^{11}	1.17 x 10^{11}	2.27 x 10^{11}	1.0 x 10^{12}	4.22 x 10^{11}

[1] *Borromeo (2007). Darlington releases include HT from the tritium removal facility.*

[2] *Brown (2005)*

[3] *CNSC (2003)*

2.4.2.4 Gas-Cooled Reactors (GCRs)

Tritium is produced by ternary fission in gas-cooled reactors (GCRs) at a rate of approximately 7 x 10^{11} Bq per MW(e)a (UN, ILO and WHO, 1983). Additional tritium on the order of approximately 7 x 10^{9} Bq per MW(e)a in liquid effluents and 10^{9} to 10^{10} Bq per MW(e)a in airborne effluents can be produced through the activation of lithium in the graphite moderator of GCRs (UNSCEAR, 1977).

Gas-cooled reactors may use either block or pebble-bed designs, referring to the packaging of fuel in either prismatic blocks or graphite-coated pebbles. Yook *et al.* (2006) compared tritium production rates for these designs. Reported rates were in the order of 1 to 3 x 10^{11} Bq per MW(e)a by ternary fission, and 4 to 9 x 10^{10} Bq per MW(e)a by activation of lithium, boron and helium, with no consistent difference in overall production among designs.

2.4.2.5 Airborne Tritium Releases from Reactors

The primary source of tritium releases to the atmosphere occurs from the reactor spent fuel pools (SFPs). Tritium builds up over time in the SFPs with age of the facility due to mixing with reactor coolant during refuelling (Jones, 2007). Mid-cycle shutdowns of the reactor result in relatively higher tritium transfer to the SFP, since the tritium inventory in the reactor coolant system (RCS) is the highest at this time. In addition, defects in the fuel cladding will further increase tritium transfer from fuel compared to diffusion alone.

The ventilation of the building that houses the SFP is designed to continuously remove tritium from the atmosphere within the building, thereby driving the evaporation rate and corresponding tritium release rate from the facility. As a result, for PWRs, the majority of tritium that is released to the atmosphere is from the SFP. In the case of HWRs, the heavy water coolant is the dominant tritium source.

2.4.2.6 Liquid Tritium Releases from Reactors

Although ion exchange is applied to chemically treat water to remove most radionuclides, thereby preventing releases to the environment, such processes are not effective at capturing tritium, since it exists primarily as HTO (Jones, 2007). As a result, in the absence of tritium removal technologies, tritium can be released to the environment during the processing of liquid waste, through system leakage and other processes (Blomeke, 1964; Haney, 1964; Jones, 2007).

Seam leaks and other primary-to-secondary coolant leaks can result in the accumulation of tritium in the secondary coolant (Jones, 2007). In addition, tritium can also naturally diffuse between the primary and secondary coolant through the U-tubes, where tritium diffusion rates tend to be relatively higher in older facilities. Steam generator blow down recovery then maintains this inventory, resulting in a build-up of tritium in the secondary coolant. However, continuous blow down to a receiving water body can result in the release of small quantities of tritium to the environment, thereby keeping the tritium inventory in the secondary coolant low.

2.4.2.7 Controlled Thermonuclear (or Fusion) Reactors

Technologies for the large-scale use of controlled thermonuclear reactors to generate heat are still under development (e.g., ITER; http://www.iter.org/). That said, if such reactors come into use, they will contain substantial inventories of tritium, posing considerable tritium management challenges (NCRP, 1979). For example, a nominal 1000 MW(e) controlled thermonuclear reactor is anticipated to produce approximately 5×10^{17} Bq per day, with an inventory of the order of 10^{19} Bq (Coyle, 1979; Crowson, 1973; Häfele *et al.*, 1977). Therefore, such reactors must be designed to prevent large releases of tritium into the environment.

2.4.2.8 Influence of Cooling Water Systems on Tritium Release

The design of the cooling water system for a reactor can influence both the location and amount of tritium released. There are two basic designs: once through cooling, and closed cycle cooling.

Once-through cooling systems use a continuous flow of lake, river or ocean water as a secondary coolant to remove waste heat from the system. With these designs, tritium as HTO in the water coolant is lost to the surface water from which that coolant was drawn. All Canadian reactors presently use once-through cooling systems.

Closed cycle cooling systems take water from lake, river or ocean only to "make up" for water lost by evaporation, wind drift or blowdown. There is very little discharge of tritium to surface water. Cooling towers or cooling ponds are used to remove waste heat from the cooling water, mainly by evaporative cooling, and tritium from the water is simultaneously lost to the atmosphere.

There are various cooling tower designs (U.S. DOE, 1993). Wet cooling towers involve direct contact between air and cooling water, and maximize the contact surface area to enhance heat exchange. Dry cooling towers do not involve direct contact; the cooling water is enclosed within a piping network, so there is no cooling water or tritium loss at the tower, but less heat exchange. Fluid coolers use a hybrid design; the cooling water does not contact the air, but clean water is sprayed on the piping and evaporated to enhance cooling. Wet cooling towers and fluid coolers can produce fog, particularly on cool, humid days, which are frequent in autumn. The fog from wet cooling towers carries tritium. On cold winter days water droplets may condense on nearby surfaces, or hoar frost may form. Tritium concentrations in condensed water will be similar to those in the recirculating cooling water.

Cooling ponds, like cooling towers, involve direct contact between air and cooling water. However, there is less exposed water surface area, less evaporation and less heat exchange. As well as releasing to the atmosphere, cooling ponds may release cooling water to surrounding soils and groundwater, and eventually to downgradient surface water.

In any closed cycle system involving evaporation of cooling water, there is a tendency for build-up of dissolved salts in the remaining re-circulated water. Cooling water may be drawn off and replaced in order to keep the salt concentration from becoming too high. Similarly, closed cycle systems will tend to accumulate higher concentrations of tritium in the re-circulating water, as compared to concentrations in a once-through system.

While there is no CANDU experience with closed cycle systems, they are not expected to significantly reduce the total losses of tritium from a facility. Rather, they would serve to redirect losses to the atmosphere, either at the cooling tower or pond, or at the condenser via air ejectors. Environmentally, their main advantage is to redirect waste heat to the atmosphere, rather than to adjacent surface waters.

2.4.3 Fuel Reprocessing Plants

Most of the tritium that has been produced in reactor fuel rods is retained within the fuel until the fuel is reprocessed (if this is done), as opposed to being released into the environment in the vicinity of the reactor site (Albenesius and Ondrejcin, 1960; Blomeke, 1964; Holmquist, 1965; UN, ILO and WHO, 1983). At the fuel reprocessing stage of the nuclear fuel cycle (if it is undertaken), uranium and plutonium are recovered from the irradiated nuclear fuel for reuse in fission reactors. During reprocessing, the uranium is first removed from its cladding material, and then dissolved in nitric acid, releasing most of the tritium into the liquid waste stream. Some tritium is also released in the dissolver off-gas and the remainder is immobilized in the cladding as solid zirconium hydride or as tritium dissolved in the metal (UN, ILO and WHO, 1983).

As is the case for reactors that use water as a primary coolant, tritium from reprocessing plants is released to the environment primarily in the form of HTO. Tritium is released to the atmosphere through stacks, as well as to the ground or to surface waters in low-level liquid wastes, where only 2 to 5% of the total fission-product tritium is retained in tanks with high-level wastes (Blomeke, 1964; Jacobs, 1968). Since there are no retention systems for tritium in the currently operating reprocessing plants, the activity released would correspond to the tritium content in the fuel elements at the time of reprocessing, with the exception of the tritium that has been immobilized in the cladding. The tritium production rate in reactors is approximately 75×10^{10} Bq per MW(e)a, and approximately half of the theoretical fuel content seems to be unaccounted for at existing tritium reprocessing plants (UN, ILO and WHO, 1983).

2.4.4 Tritium Production Facilities

Because little tritium is naturally present, it must be produced artificially for use on a practical scale. To accomplish this, tritium can be generated in production reactors that have been specifically designed to optimize the production of tritium and other nuclear materials (ANL, 2005). In such facilities, tritium can be produced through irradiation of lithium metal, alloys or salts (General Nuclear Engineering Corp., 1959; Jacobs, 1968; Johnson, 1962; Magee *et al.*, 1960). For example, a ^{6}Li atom can combine with a neutron in the process of neutron activation to form a ^{7}Li atom. The ^{7}Li atom then immediately splits to form tritium and ^{4}He (as shown in Equation 2.5 above). This tritium is isotopically separated from other hydrogen isotopes and subsequently processed in a tritium-handling plant (Crowson, 1973).

Based on data for the Savannah River Plant (the primary source of tritium in the United States), airborne emissions via the stacks occur at a rate of 4.1×10^{16} Bq/a on average, and can range from 1.4×10^{16} to 9.9×10^{16} Bq/a (Murphy and Pendergast, 1979). Releases from such facilities consist of approximately 20% HT and 80% HTO under normal operating conditions, with accidental releases primarily in the form of gaseous HT.

Accidental airborne releases can raise the contribution of HT to values on the order of 60% of the total tritium discharged (Murphy and Pendergast, 1979). By comparison, the amount of tritium released in the liquid effluents seems to represent approximately 10% of the airborne releases (NCRP, 1979). Releases from tritium production facilities can be reduced through the implementation of measures to optimize tritium yields during processing (Jacobs, 1968).

Tritium can also be produced in accelerators by bombarding ^{3}He with neutrons (as depicted in Figure 2.4), although this approach has not been applied on a large scale (Zerriffi, 1996). Tritium can be formed in the light water cooling of electron or proton accelerators primarily through spallation of the oxygen atoms in the water molecules. Small amounts of tritium can also be produced through spallation of nitrogen, carbon and other light molecules. It is possible that some releases of this tritium to the environment can occur through airborne releases and possibly liquid releases depending upon the design of the cooling system.

2.4.5 Consumer Products

The fractional release of tritium, in the form of HTO, HT and short-chain organic radicals of the styrene type, from luminous compounds (such as those used as dial paints for the illumination of timepieces), comprises approximately 5% annually (Comps and Doda, 1979; Krejci, 1979; Krejci and Zeller, 1979; UNSCEAR, 1977; Wehner, 1979). Such releases can represent amounts on the order of 10^{14} Bq tritium per year (Krejci, 1979).

Tritium releases from gas-filled tubes, such at those used in liquid crystal displays (LCDs) and inside exit signs or electronic tubes are negligible in comparison, showing values of approximately 2×10^{12} Bq tritium per year (Krejci, 1979; UN, ILO and WHO, 1983). That said, however, environmental releases following accidental breakage of tubes or improper disposal can be significant (Comps and Doda, 1979; Wehner, 1979).

Mutch and Mahony (2008) reported tritium levels in municipal landfill leachates, which were attributed to disposal of such products. In a study of ten landfills in New York and New Jersey, the average tritium as HTO in leachate was 1,251 Bq/L and the maximum was 7,104 Bq/L. A similar study of California landfills indicated an average tritium in leachate of 3,663 Bq/L and a maximum of 11,248 Bq/L. Landfill gas condensates in U.K. and California studies had tritium concentrations as high as 2,013 Bq/L and 18, 981 Bq/L, respectively.

3.0 PHYSICAL AND CHEMICAL BEHAVIOUR OF TRITIUM IN THE ATMOSPHERE

This section outlines the physical behaviour of tritium in the atmosphere, the chemical transformations of tritium that occur in the atmosphere, factors important to atmospheric dispersion of tritium plumes, and possible climate change effects on tritium behaviour and environmental transport.

3.1 Physical Behaviour of Tritium

3.1.1 Radioactive Decay

Tritium is the only radioactive isotope of hydrogen that has been observed in nature, although extremely short-lived radioisotopes of hydrogen (i.e., ^{4}H to ^{7}H) have been produced under laboratory conditions (Audi and Wapstra, 1995; Friedlander *et al.*, 1981; Kelley and Tilley, 1987; Kinsey, 1999). Tritium is a pure beta-emitter with a half-life of 12.32 years, a maximum energy of 18 keV and a mean energy of 5.7 keV (Lucas and Unterweger, 2000; UN, ILO and WHO, 1983; Unterweger *et al.*, 1980). Tritium decays to produce a stable ^{3}He daughter, as described by the following equation:

$$^{3}H \rightarrow {}^{3}He + \beta^{-} + \text{Energy} \qquad \text{Equation 3.1}$$

3.1.2 Other Physical Properties

Tritium transfer in the environment is driven by a number of physical processes. These include isotopic replacement (ie. isotopic exchange) and diffusion.

Diffusion is the mixing of tritium within and between environmental compartments as a result of random molecular motion which transports matter from one point to another. Since molecules are in constant motion, diffusion occurs in gases, liquids and solids. For example, gaseous HT diffuses into soil pore spaces, where it is rapidly oxidized to HTO by microorganisms (12% to 66% per hour depending on soil type) (Dunstall *et al.*, 1985a,b; McFarlane *et al.*, 1978, 1979). In general, the rate of diffusion will increase with increasing temperature and turbulence in a given environmental phase, thereby increasing the rate of transport and mixing in the environment. This can lead to seasonal changes in environmental transport of contaminants, such as tritium, by the diffusion process.

In addition to diffusion, further transport of tritium in the environment can occur through the process of isotopic replacement. Isotopic replacement is a process by which two atoms representing different isotopes of the same element exchange locations in the same molecule or different molecules. For example, in tritiated water, it occurs when a tritium atom from one water molecule changes places with a hydrogen atom from an adjacent water molecule (ANL, 2005). As a result, tritium is naturally present as a very small percentage of ordinary hydrogen in water, both in the liquid and gaseous states. Isotopic replacement of hydrogen

atoms by tritium can also occur for tritium of anthropogenic origin, as well as for molecules other than water that contain hydrogen atoms. For example, at room temperature, the combination of hydrogen gas (H_2) and T_2 to produce HT is favoured (Jones, 1948).

Isotopes of the same element take part in the same chemical reactions, but because the atoms of different isotopes differ in terms of their size and atomic weight, they tend to react at different rates. For example, physical processes, such as evaporation and diffusion, can discriminate against heavy isotopes because they move more slowly in the environment. In addition, enzymatic discrimination and differences in kinetic characteristics and equilibria can result in reaction products that are isotopically heavier or lighter than their precursor materials. In the case of hydrogen isotopes, although tritium closely follows the reactions of ordinary hydrogen, the relatively large mass differences between the hydrogen isotopes (as shown in Table 3.1) make isotopic effects easily discernible (Jacobs, 1968).

Table 3.1: Atomic Weights of Different Hydrogen Isotopes Contributing to Isotopic Fractionation in the Environment (from Lange and Forker, 1961).

Hydrogen Isotope	Symbol	Atomic Weight (g)
Proton	^1H	1.00814 ± 0.000003
Deuteron	^2H	2.014735 ± 0.000006
Triton	^3H	3.016997 ± 0.000011

As discussed above, physical processes, such as phase changes, that can influence tritium transport in the environment are expected to be slower for heavier isotopes. For example, Table 3.2 provides a summary of some key thermodynamic properties for the different oxides of hydrogen isotopes, including H_2O, D_2O and T_2O (Bigeleisen, 1962; Jacobs, 1968; Smith and Fitch, 1963). Based on these data, increases in boiling point and heat of vaporization can be discerned with increasing atomic weight of the hydrogen isotopes. Such physical differences between hydrogen isotopes would be expected to lead to corresponding declines in the rates of evaporation in nature for isotopically heavier water molecules that have relatively higher boiling points and heats of vaporization. In addition, isotopes with higher entropies could lead to increased rates of mixing.

Table 3.2: Thermodynamic Properties of the Oxides of Hydrogen Isotopes (Bigeleisen, 1962; Jacobs, 1968; Smith and Fitch, 1963).

Thermodynamic Property	Oxide of Hydrogen Isotope		
	H_2O	D_2O	T_2O
Boiling point (°C)	100.00	101.42	101.51
Triple-point temperature (°C)	0.010	3.82	4.49
Triple-point pressure (mmHg)	4.58	5.02	4.87
Heat of vaporization at the boiling point (kcal/mole)	9.72	9.9	10.1
Entropy (at 298 K)	16.75	18.9	19.0

Physical conditions in the environment, such as temperature, pressure, relative humidity, and the presence and intensity of a concentration gradient in the environment, can also influence tritium transport. For example, both reaction rates (including rates of isotopic replacement, oxidation from HT to HTO, biological activity, etc.) and rates of molecular motion (which can increase the rate of environmental mixing) are expected to increase with temperature. In addition, increases in temperature can result in phase changes (e.g., from solid to liquid and from liquid to vapour). Decreases in temperature can result in opposite phase changes. The solubility of a solid in water will generally increase with increasing temperature, while the solubility of a gas in water will decrease with increasing temperature. Increases in pressure can also lead to increases in reactivity rates, as well as physical mixing due to molecular motion, which increases with pressure.

The phase change from water to water vapour is of particular interest. Vapour is the name given to gaseous molecules of a substance at a temperature and pressure at which we normally think of the substance as a liquid. Liquids with weak molecular attractive forces, such as water, vaporize readily at temperatures well below the boiling point. The opposite process, when vapour molecules coalesce into droplets and adhere to the water surface (or another surface) is called condensation. The vapour pressure is the pressure of the vapour in a closed space over the liquid, i.e., at saturation, when vaporization and condensation rates are equal. Vapour pressure increases with temperature.

A sudden drop in temperature under humid conditions may cause water vapour in air to condense into fine droplets known as fog. The dew point is the temperature to which air must be cooled to trigger condensation of its water vapour. If the dew point is below the freezing point it is often called the frost point, because condensation will form hoar frost. The dew point (T_d) can be roughly calculated from temperature (T) and relative humidity (H) as follows (Lawrence, 2005):

$$T_d = T - (100 - RH)/5 \qquad \text{Equation 3.2}$$

This equation is accurate within ±1°C when relative humidity is greater than 50%. Autumn weather is particularly conducive to fog formation. Release of warm, humid stack gases to cooler ambient air may also produce localized fog, in the form of a visible vapour plume.

3.1.3 Mobility in the Environment

As discussed in Section 2.0 above, tritium behaves much like hydrogen in the environment, except for its decay to helium, as well as some isotopic discrimination between hydrogen isotopes (Section 3.1.2 above). Natural and fallout tritium are primarily produced in the stratosphere, where they occur in the form of tritiated water (or HTO) for the most part (UN, ILO and WHO, 1983).

Altitude has a pronounced effect on the reactions that occur in the atmosphere, with the tendency for TO_2 to be consumed to form HTO at altitudes of below 5 km (Jacobs, 1968). By comparison, at altitudes between 10 and 40 km, HT is expected to be the predominant form of tritium, whereas above 40 km, the small concentrations result in a negligible

contribution of HT to total tritium levels (Harteck, 1954). The tritium concentration in atmospheric hydrogen is expected to remain relatively stable, whereas localized variability in rainwater tritium levels may occur (Harteck, 1954; Libby, 1953).

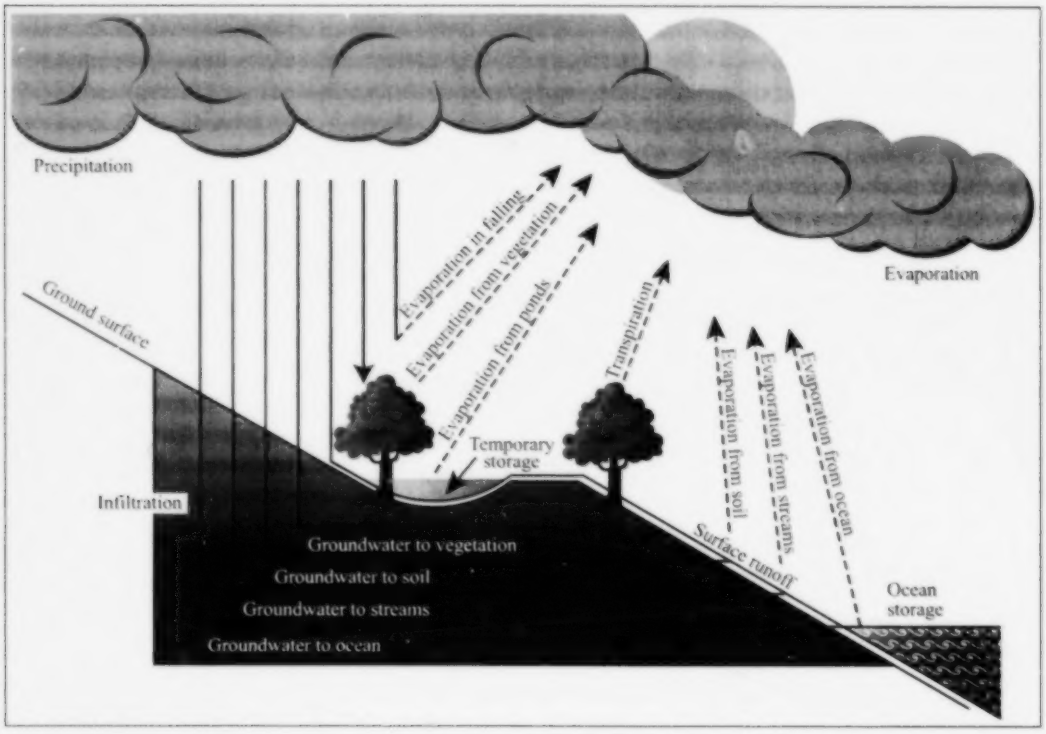
In thermonuclear clouds, tritiated hydrogen is produced in the stratosphere by radiative dissociation of water under oxidizing conditions (Martell, 1963). Tritiated methane is formed through tropospheric exchange of methane with the tritiated hydrogen that has moved down from the stratosphere. The tritium specific activity of atmospheric methane is several orders of magnitude higher than that of rainfall, but somewhat less than that of atmospheric hydrogen and stratospheric water vapour (Harteck, 1954).

Following production in the upper atmosphere by cosmic radiation, tritium atoms have a high kinetic energy and are readily oxidized (Jacobs, 1968). At lower than atmospheric pressures, this likely predominantly occurs through a three-body collision with oxygen (O_2) to initially form a relatively stable radical, HO_2 (Harbeck, 1954). This radical can then react with ozone (O_3) following repeated photochemical decomposition reactions of TO_2 to form HTO, as opposed to HT, in the atmosphere. Once tritium has been incorporated into water molecules, HTO then falls to the surface of the earth as rain and snow, entering the natural hydrological cycle in this way (Figure 3.1; Stamoulis *et al.*, 2005; UN, ILO and WHO, 1983).

A second feasible initial reaction would involve the collision of tritium with an H_2 molecule, with a resultant isotopic exchange to form HT. Small amounts of tritium, representing less than 1% of the amount produced per day, may also be produced through the initiation of a reaction between tritium and oxygen by the beta radiation from tritium decay (Dorfman and Hemmer, 1954; Yang and Gevantman, 1960).

Transfer of tritiated water vapour from the stratosphere to the troposphere occurs with a half-time of approximately a year, after which it is deposited onto the earth's surface as rainfall and through molecular exchange with a half-time on the order of 10 days. Deposited HTO is then transported in the environment as part of the hydrological cycle (Figure 3.1; Jacobs, 1968).

Figure 3.1: Conceptual Model Depicting the Hydrological Cycle (from Baver, 1956).

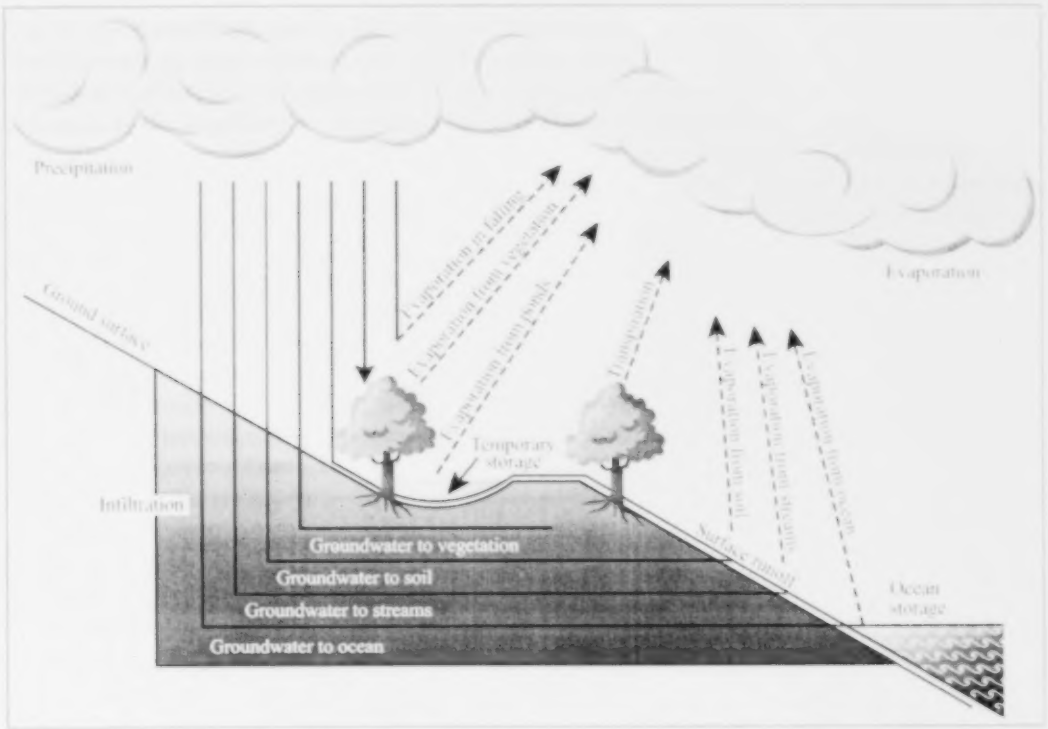


Following deposition onto the ocean surfaces, tritiated water becomes diluted in the mixed layer and later, a proportion of this HTO evaporates back into the atmosphere at a much lower concentration. A smaller fraction is transported to the deep ocean (UN, ILO and WHO, 1983). By comparison, tritiated water that has been deposited onto land surfaces partitions between surface run-off to ponds, lakes, streams or oceans and infiltration into the soil. Following soil infiltration, HTO can be absorbed by plants, can evaporate and re-enter the atmosphere, or can be carried below the surface via the groundwater to fresh water bodies on the land surface or to the ocean.

Most of the tritium released to the lower atmosphere can be found in the ocean within a year (NCRP, 1979). A small amount (21%) enters soil water, fresh surface water and groundwater, and takes a longer route to the ocean. After 70 years, less than 9% is in groundwater and the remainder is in the ocean.

Tritium levels in continental surface waters (as HTO) tend to be higher than those in the ocean surfaces (Brown, 1964; Eriksson, 1965; Israel *et al.*, 1963, Schell *et al.*, 1974; Weiss *et al.*, 1979b, Momoshima *et al.*, 1991). This is due to the relatively small dilution following mixing with surface water over land masses, compared to the potential for dilution in oceanic waters. As a result, tritium activity concentrations in water vapour that has undergone

Figure 3.1: Conceptual Model Depicting the Hydrological Cycle (from Baver, 1956).



Following deposition onto the ocean surfaces, tritiated water becomes diluted in the mixed layer and later, a proportion of this HTO evaporates back into the atmosphere at a much lower concentration. A smaller fraction is transported to the deep ocean (UN, IEO and WHO, 1983). By comparison, tritiated water that has been deposited onto land surfaces partitions between surface run-off to ponds, lakes, streams or oceans and infiltration into the soil. Following soil infiltration, HTO can be absorbed by plants, can evaporate and re-enter the atmosphere, or can be carried below the surface via the groundwater to fresh water bodies on the land surface or to the ocean.

Most of the tritium released to the lower atmosphere can be found in the ocean within a year (NCRP, 1979). A small amount (21%) enters soil water, fresh surface water and groundwater, and takes a longer route to the ocean. After 70 years, less than 9% is in groundwater and the remainder is in the ocean.

Tritium levels in continental surface waters (as HTO) tend to be higher than those in the ocean surfaces (Brown, 1964; Eriksson, 1965; Israel *et al.*, 1963; Schell *et al.*, 1974; Weiss *et al.*, 1979b; Momoshima *et al.*, 1991). This is due to the relatively small dilution following mixing with surface water over land masses, compared to the potential for dilution in oceanic waters. As a result, tritium activity concentrations in water vapour that has undergone

re-evaporation from continental water bodies are similar to those occurring in the original precipitation, particularly for small water bodies. By comparison, precipitation depositing on the ocean or on other deep water bodies is rapidly mixed, and greatly diluted, resulting in lower tritium concentrations in re-evaporated water vapour compared to the levels in the original precipitation.

Concentrations of tritium in the atmosphere are also known to vary across latitudes and seasons (e.g., Hauglustaine and Ehhalt, 2002). For example, in addition to the observed difference in continental water versus oceanic tritium levels, differences also occur as dictated by the latitudinal movement of air masses. Specifically, tritium concentrations are typically higher in precipitation originating from air masses with trajectories from the north and west, compared to those originating from the south and east, likely primarily due to latitudinal differences in tritium transfer rates from the stratosphere (Brown, 1964).

Vapour exchange processes are thought to account for approximately two-thirds of the tritium removal into the oceans (Erikkson, 1965), and also account for much of the tritium transfer to terrestrial soil water. Additional exchange can occur between atmospheric tritium and precipitation, as dictated by rainfall intensity, as well as the size and corresponding surface area of precipitation droplets. Precipitation then transfers tritium to the earth's surface.

In the case of light rainfall, tritium levels in precipitation are thought to be representative of those occurring at lower heights in the atmosphere, whereas for moderate to heavy rains, exchange at the lower levels is considered to be insignificant (Bolin, 1961). As noted by Sejkora (2006), light, long-duration rainfall events can yield higher tritium concentrations in the ground water, runoff, storm drains and soils than heavy, short-duration events. This may be important when considering the influence of severe storm events and the potential influence of climate change on this phenomenon.

Tritium concentrations in precipitation (as HTO) have been found to increase with length of the travel path through the air mass. For example, comparisons of tritium levels in precipitation with meteorological factors (such as type of storm, radar height of precipitation formation, rainfall duration and intensity, and the type and origin of the air mass) have revealed increases in precipitation tritium levels as the depth of the air mass being traversed by the precipitation increases (Brown, 1964).

Related to the intensity of the precipitation, the size and surface area of the rain droplets can also influence tritium exchange between water vapour and rain drops during the process of film diffusion across the droplets (Chamberlain and Eggleton, 1964). In general, the tritium specific activity of rain drops that have passed through a plume that is contaminated with tritium is very dependent upon the size of the droplet, with decreases in tritium concentrations in larger rain drops due to the smaller surface area-to-volume ratio of larger droplets. This essentially leads to relatively greater tritium dilution in larger droplets.

In the case of snow and ice particles, it is expected that tritium would exchange at the air-to-particle interface on particle surfaces. However, such processes may be less pronounced for snow and ice due to the lower temperatures, as well as the slower rates of diffusion into frozen particles compared to rain droplets.

Davis (1997) and Konig *et al.*, (1984) have determined washout coefficients during snowfall, and Davis has contrasted those with washout coefficients during rainfall. The washout coefficient is a rate constant (s^{-1}) that reflects the rate of tritium scavenging from the air by snowfall or rainfall. For a 1 mm/h (water equivalent) snowfall, washout coefficients of 2.1×10^{-5} s^{-1} and 2.6×10^{-5} s^{-1} were reported by Davis (1997) and Konig *et al.*, (1984), respectively. In contrast, higher washout coefficients of approximately 10^{-4} s^{-1} have been reported for rainfall events of similar intensity (Chamberlain and Eggleton, 1964; Tadmor, 1973; Abrol, 1990; Gulden and Raskob, 1992).

Dry deposition of tritium to the snowpack in winter or to soil water in summer occurs by direct vapour exchange with snow or soil water. This transfer is often expressed as a deposition velocity (m/s). Davis (1997) determined a deposition velocity of 1.6×10^{-3} m/s for tritium transfer to the winter snowpack. Barry (1964) reported a winter deposition velocity 3.0×10^{-3} m/s. In contrast, higher deposition velocities of 4.0×10^{-3} m/s and 2.0×10^{-3} m/s have been reported by Gulden *et al.* (1990) and Bunnenberg *et al.* (1992) under summer conditions.

Seasonal factors also influence the timing of tritium transport between environmental compartments. For example, following deposition onto the ground surface, tritiated rain (in its liquid state) can enter groundwater and surface waters through infiltration and runoff, respectively, during periods when the ground is not frozen or covered with snow. In addition, evaporation of aqueous HTO to the air can occur. By comparison, during the winter, if the ground is snow-covered, tritium can deposit and accumulate in the snowpack, where it typically remains until it is released during snowmelt (Davis, 1997). This can lead to seasonal pulse inputs of tritium to soil and water bodies during thaws. Further delays in tritium release from the snowpack to the atmosphere can occur due to the slower tritium exchange rates that would be expected between air and snow during the winter.

As an example of snowmelt effects, studies at the SRBT facility (Section 5.1.1) show that tritium is elevated in spring snow melt samples collected at the north-west corner of the property in the vicinity of the stack which releases tritium to the atmosphere. Snow storage in this area probably contributes to elevated concentrations of tritium in groundwater at this location.

3.2 Chemical Transformations of Tritium

Industrial tritium releases predominantly consist of HTO and HT, and under some conditions, tritiated methane (CH_3T) (ATSDR, 2002; McCubbin *et al.*, 2001). The residence times of HT and CH_3T in the atmosphere are not well known, although mean values of 5 to 10 years have been estimated (e.g., Burger, 1979). Of the possible anthropogenic forms of tritium, HTO is of most interest, since it tends to be much more biologically-active than other forms (UN, ILO and WHO, 1983).

When tritium is released as a gas (HT) or as HTO in its vapour form (such as from airborne releases), it can be concentrated by ordinary chemical separation of hydrogen and water vapour from air. In contrast, when present as HTO in aqueous solutions (such as in liquid effluents), it is not amenable to separation and concentration by conventional waste-treatment techniques (Blomeke, 1964; Jacobs, 1968). Instead, isotopic separations are required, which may be difficult. The more satisfactory procedures of isotopic separation are usually physical in nature because mass differences result in greater relative differences in physical properties than in chemical properties (Moeller, 1954). As a result of the similarity of the properties of HTO and H_2O, tritium generally follows ordinary water in processing streams and in the environment under such conditions (Haney, 1964; Moeller, 1954).

3.2.1 Tritium Transformation from HT to HTO

Upon release into the atmosphere, key removal processes for HT include photochemical oxidation in air and bacterial oxidation in soil, resulting in transformation to HTO (Dunstall *et al.*, 1985a, 1985b; Hart, 2008). The atmospheric process is slow, with a half-life of > 5 years, while a much higher oxidation rate occurs at the air-soil interface with a half-life of 40 minutes to 5 hours (Mishima and Steelec, 2002; McFarlane *et al.*, 1978, 1979). The chemical reactions are:

$H_2 + OH \longrightarrow H + H_2O$	*photochemical*	Equation 3.3
$H + O_0 + M \longrightarrow HO_2 + M$	*(M =other molecule)*	
$2H_2 + O_2 \longrightarrow H_2O$	*bacterial*	

Some of the HTO that is formed at the air-soil interface is taken up by plants through their roots with transpiration water, and some is emitted to the atmosphere (Neil, 1991, 1992). This HTO is available for uptake by animals and humans through inhalation and ingestion, as well as by plants through their leaves.

$$P_{11a} = R_{HT} \bullet H_a \bullet f_{oxid} \qquad \text{Equation 3.4}$$

where: P_{11a} = transfer parameter from HT in air to HTO in air ($Bq \bullet m^{-3}/Bq \bullet m^{-3}$)

R_{HT} = ratio of HTO concentration in air moisture ($Bq \bullet L^{-1}$) to HT concentration in air ($Bq \bullet m^{-3}$)

f_{oxid} = fraction of year when oxidation can occur (unitless)

H_a = absolute humidity ($L \bullet m^{-3}$)

Default values of P_{11a}, as recommended by Hart (2008), have been provided in Table 3.3.

Table 3.3: Regional Default Values for P_{11a}, Transfer from HT to HTO in Air (Unitless).

Parameter	Southern Ontario	Western Ontario	Eastern Ontario	Quebec	Maritimes
H^a (snow free) ($L \bullet m^{-3}$)	0.0089	0.0085	0.0080	0.0076	0.0075
f_{oxid} (unitless)	0.75	0.75	0.67	0.67	0.75
P_{11a} (unitless)	0.053	0.051	0.043	0.041	0.045

Notes:

- *RHT [$Bq \bullet L^{-1}(HTO) \bullet Bq^{-1} \bullet m^3(HT)$] = 8*
- *Site-specific data for CRL indicates R_{HT} = 4, but this may not apply to all of Eastern Ontario; therefore default is 8.*

The best estimate for R_{HT} is 4 $Bq \bullet L^{-1}$ HTO per $Bq \bullet m^{-3}$ HT as determined in the 1994 HT chronic release experiment at Atomic Energy of Canada Limited (AECL)'s Chalk River Laboratories (CRL) (Davis *et al.*, 1995a,b; Davis and Bickel, 2000; Spencer *et al.*, 1996). This value is a factor of 2 higher than estimated by Neil (1992) from indirect evidence. It is uncertain because the HTO concentrations measured in the field were very close to background levels and because the concentrations may not have reached steady state by the end of the study period. Also, the value may not be directly applicable to the other nuclear facility sites if soils there are significantly different than those at CRL in terms of the properties that control HT deposition and oxidation (water content, soil porosity and distribution of microorganisms). Moreover, there is little information indicating how R_{HT} values are distributed. A lognormal distribution is assumed since many parameters expressed as ratios are log normally distributed and since Neil (1992) shows a distribution that is roughly lognormal. Accordingly, the value 4 $Bq \bullet L^{-1}/(Bq \bullet m^{-3})$ is taken as the GM for R_{HT} and the GSD is set by judgement at 1.5, implying a 95% confidence interval ranging from about 1.8 to 9 $Bq \bullet L^{-1}/(Bq \bullet m^{-3})$. A value of 4 is recommended as a default for sites with soils similar to those at CRL (sandy loam); for other soil types a value of 8 $Bq \bullet L^{-1}/(Bq \bullet m^{-3})$, which is believed to be conservative in all situations (Davis and Bickel, 2000), should be used.

The absolute humidity, H_a, should be assigned a site-specific value for use in Equation 3.2. H_a is not normally measured directly, but can often be derived from the amount of water extracted from the molecular sieve, and the volume of air sampled, in site monitoring

programs for tritium concentrations in air (Hart, 2008). In the absence of local data, default regional values are given in Table 3.4 for different seasons of the year. The values vary significantly with averaging time and must be chosen to match the way in which H_a appears in a model. For example, an average over the snow-free period is appropriate for calculating the conversion of HT in air to HTO in air (Equation 1.8). Shorter averaging times are more appropriate for H_a used in modeling conversion of HT in air to HTO in plants.

The factor, f_{oxid}, is the fraction of the year when the soil is not frozen or snow covered, and is applied to allow for the decrease in HT oxidation and HTO re-emission rates in the winter. Site-specific data for this parameter should be used if available. For Ontario in the Chalk River/Pembroke area and for Quebec near Gentilly-2 (G-2), the recommended value for f_{oxid} is 0.67, based on the frost-free period reported in the Environment Canada Climate normal database for nearby climate stations. For sites in southern Ontario (e.g., near Bruce Power (BP), the Pickering Nuclear Generating Station (PNGS) and the Darlington NGS (DNGS)) and for Point Lepreau, f_{oxid} should be assigned a value of 0.75.

Table 3.4: Absolute Humidity, H_a ($L \cdot m^{-3}$) Averaged over Various Seasons of the Year at Stations Close to Canadian CANDU Facilities (from Hart, 2008).

Site	Annual Average	Average Over the Snow-free Period	Average Over the Growing Season
Toronto Island (Pickering)	0.0069	0.0089	0.012
Trenton (Darlington)	0.0069	0.0089	0.012
Wiarton (Bruce)	0.0066	0.0085	0.011
Ottawa (CRL)	0.0050	0.0080	0.010
Quebec City (Gentilly-2)	0.0047	0.0076	0.010
Saint John (Point Lepreau)	0.0054	0.0075	0.010

3.2.2 Soil Oxidation of Tritiated CH_4

The methane content (CH_4) of the atmosphere is rather high, compared to other trace gases. The CH_4 is strongly coupled with the H_2 and CO cycles, contributing approximately 1% to the total atmospheric carbon cycle, as estimated by Ehhalt (1974).

Oxidation of CH_4 leads to the formation of carbon dioxide (CO_2) and water. The large majority of the methane oxidation takes place in the upper atmosphere by photochemical reactions with OH (Burger, 1979; Freyer, 1977; Hart, 2008), with some methane oxidation by methanotrophic microorganisms in soils under aerobic conditions (Mancinelli, 1995), as described by the following equation:

$$CH_4 + 2\,O_2 \longrightarrow CO_2 + 2\,H_2O \qquad \text{Equation 3.5}$$

Isotopic exchange between tritium and ordinary hydrogen atoms of methane can produce tritiated methane, and this in turn can be transformed to HTO. In the reverse process that is carried out by methanogenic bacteria under anaerobic conditions (Zehnder and Brock, 1979), HTO can be converted back to tritiated methane.

Methane oxidation can also occur through the reduction of sulphate by microorganisms during anaerobic metabolism (Mancinelli, 1995), as described by the following equation:

$$2CH_4 + SO_4^{2-} \longrightarrow 4H_2 + 2CO_2 + H_2S \qquad \text{Equation 3.6}$$

Anaerobic sulphate reduction has only been found to occur in environments where sulphate is present, and tends to predominate in aquatic systems (Mancinelli, 1995). Through this process, tritiated methane can be converted to HT and HTS gas.

During methane formation and oxidation, it is expected that the heavier tritium atoms would react more slowly and would bind more strongly than the lighter hydrogen isotopes, leading to isotopic fractionation by soil microorganisms.

Topp and Pattey (1997), Born *et al.* (1990) and Crutzen (1991) indicate that an average of about 7% (range of 1 to 15%) of the methane released globally is oxidized at ground level by micro-organisms in soil, with the subsequent formation of water (and in some cases, HTO), as described by Equation 3.4 above. Since these values represent very broad averages, and there is no accounting for proximity to the source of CH^4, local oxidation fractions are likely to be less. Kim *et al.* (2004) show that the oxidation fractions within 10 km of the source are 0.24% for Pickering and Darlington, and 0.1% for Bruce Power, G2 and Point Lepreau. On this basis, an oxidation fraction equivalent to 0.3% can be conservatively assumed to apply to local ground-level oxidation of CH_4.

3.3 Tritium Behaviour in Atmospheric Plumes

3.3.1 Relevant Forms of Tritium

Tritium can exist in various forms in atmospheric plumes from nuclear facilities, the most important being tritiated water (HTO) and tritiated hydrogen gas (HT). The relative proportion of these various forms of tritium in air around a facility depends on the proportions released from the facility, and also on atmospheric chemistry processes, the most important being the water vapour content of air (Momoshima *et al.*, 2007). Because water vapour saturation is temperature dependant, the water vapour content and consequently the conversion of HT to HTO (see Section 3.2) will be higher during the summer than in winter.

The form of tritium is important to both environmental partitioning and radiological dose imparted to people living around a facility. Gaseous HT reacts more slowly than HTO with other environmental compartments, and imparts a very low radiological dose relative to HTO because it is only weakly absorbed by the body. The HTO behaves like water in the

atmosphere, partitions easily to soil and plants, and imparts a larger radiological dose in the body. As noted by Spencer and Vereecken-Sheehan (1994), HTO is usually the more abundant chemical form in plumes arising from nuclear facilities.

3.3.2. Characteristics of Tritium Releases at Nuclear Facilities

In addition to the type of tritium released, the atmospheric dispersion of contaminants depends on many facility-specific factors such as number of release stacks, stack height, exhaust velocity, gas exit temperature, air moisture (humidity), size and location of adjacent buildings, local atmospheric turbulence and wind conditions, and local topography. It is important to understand whether stack emissions are continuous or intermittent and whether the amounts released are constant or variable. Since these factors vary on a facility-by facility basis it is difficult to generalize about their relative importance.

The specific release characteristics are more important when considering short-term releases and trying to predict air concentrations for this period. They can be particularly important for receptors located in the near-field. However, they are less important when considering long-term annual average concentrations, which are primarily of interest in determining facility impacts associated with tritium releases to air. A further discussion of how facility specific factors can affect the atmospheric dispersion of tritium is provided in the following section.

3.3.3 Dispersion of Tritium Plumes

3.3.3.1 Normal Dispersion Processes

Tritium plumes undergo similar dispersion processes to other types of contaminant plumes. Releases of HT or HTO disperse in similar fashion. The plume rises from the stack until it is bent over by the wind, after which it travels downwind, spreading horizontally and vertically as it goes. The final rise of the plume depends on atmospheric conditions and on the characteristics of the release (stack diameter, exit velocity, temperature). Generally, the rise is highest under low-wind conditions and lowest under strong winds.

The horizontal and vertical spread or dispersion of the plume depends on atmospheric conditions. The dispersion tends to be greatest under calm, clear, daytime conditions and least under calm, clear, nighttime conditions. The former is referred to as unstable or convective conditions and the latter is known as stable or inversion conditions. Moderate and high wind conditions result in an intermediate level of dispersion. This condition, which is known as neutral stability, is the most common and tends to have a dominant effect on long-term average plume concentrations.

The lateral profile of concentration within a dispersing plume approximates a symmetric, bell-shaped curve, which is known as the Gaussian distribution. The Gaussian distribution is the basis of the various numerical dispersion models used to predict the dispersion of contaminant plumes. The calculation of the spread rate for the Gaussian distribution, which

depends on the atmospheric stability, has been largely based on the work of Pasquill (1962). In the vertical direction, the Gaussian distribution has proven to be a good approximation for neutral conditions, but not as good for stable and unstable conditions. A relatively recent innovation in dispersion models is to replace the Gaussian distribution with a more sophisticated distribution for these conditions. Another relatively recent innovation is to use more sophisticated approaches for estimating the spread rate as a function of atmospheric stability and terrain conditions. The United States Environmental Protection Agency's (US EPA's) newest generation of dispersion model, AERMOD, incorporates these effects.

The dispersion model currently recommended in Canada to assess long-term exposure from tritium releases at nuclear facilities is outlined in the N288.1-08 Guideline (CSA, 2008). Like most dispersion models, this one is based on the Gaussian distribution. It calculates long-term airborne concentrations using a sector-averaging approach, similar to that incorporated into the Long-Term version of the US EPA's Industrial Source Complex model (ISC-LT), which is a predecessor of AERMOD. In this approach, meteorological data for the study site are categorized into 16 wind direction sectors and analyzed to determine frequencies of occurrence for various categories of wind speed and atmospheric stability. This information is then used to calculate weighted average dispersion and ground-level concentration by sector.

The sector-averaged Gaussian plume model is formulated in N288.1-08 as a dispersion or transfer factor $(P_{01})_j$, which is multiplied by the tritium release rate to give the tritium concentration in ground-level air in wind sector j at distance x from the point of release. Mathematically, $(P_{01})_j$ is calculated as follows:

$$(P_{01})_j = [(2/\pi)^{1/2} /(x\,\Delta\theta)] \sum [F_{ijk}\, D_k \exp(-H_{ik}^2 / 2\Sigma_{zi}^2) / (u_k\, \Sigma_{zi})]\,, \qquad \text{Equation 3.7}$$

where $(P_{01})_j$ is the ground-level transfer factor for receptor j ($s \cdot m^{-3}$),
x is the distance between the source and receptor j (m),
$\Delta\theta$ is the width of the sector over which the plume spreads (radians),
F_{ijk} is the triple joint frequency of occurrence of stability class i and wind speed class k when the wind blows into the sector containing receptor j,
D_k is a factor that takes account of decay and ingrowth for wind speed class k,
H_{ik} is the effective release height for stability class i and wind speed class k (m),
Σ_{zi} is the vertical dispersion parameter for stability class i, including spreading due to building wake effects (m), where z refers to the vertical axis, and
u_k is the mean wind speed for speed class k ($m \cdot s^{-1}$).

The effective release height H_{ik} is the physical stack height, adjusted if necessary for the effects of plume rise due to thermal buoyancy or momentum (Section 3.3.3.3) and for the effects of stack or building wakes (Section 3.3.3.4). The N288.1-08 Guideline provides the detailed adjustment procedures.

The decay and ingrowth term D_k reduces to a simple decay term for tritium, since there are no radioactive progeny of interest. Thus, $D_k=\exp(-\lambda \cdot (x/u_k))$, where λ is the decay constant of 1.79×10^{-9} s^{-1}. The travel time to distance x is x/uk. Since receptors of interest are generally within a few km, at typical wind speeds of about 2 m/s, tritium will reside for approximately 30 min in the area of interest.

With modern day computational power, more detailed models that determine long-term concentrations by performing dispersion calculations for each hour of the meteorological record are now readily available, such as in AERMOD. As previously mentioned, AERMOD also provides more refined approaches for predicting dispersion under unstable and stable atmospheric conditions. However, since long-term exposure tends to be dominated by neutral rather than stable or unstable conditions, these refinements have only a modest effect on the prediction of long-term concentrations.

The available evidence from monitoring at Canadian nuclear facilities (Chouhan and Davis, 2001) shows that the sector-averaged Gaussian model works reasonably well, predicting annual average tritium within a factor of 2, with a slightly high bias on average. Atmospheric processes that may influence tritium dispersion in defined situations are discussed in the following sections. They are illustrated in Figure 3.2.

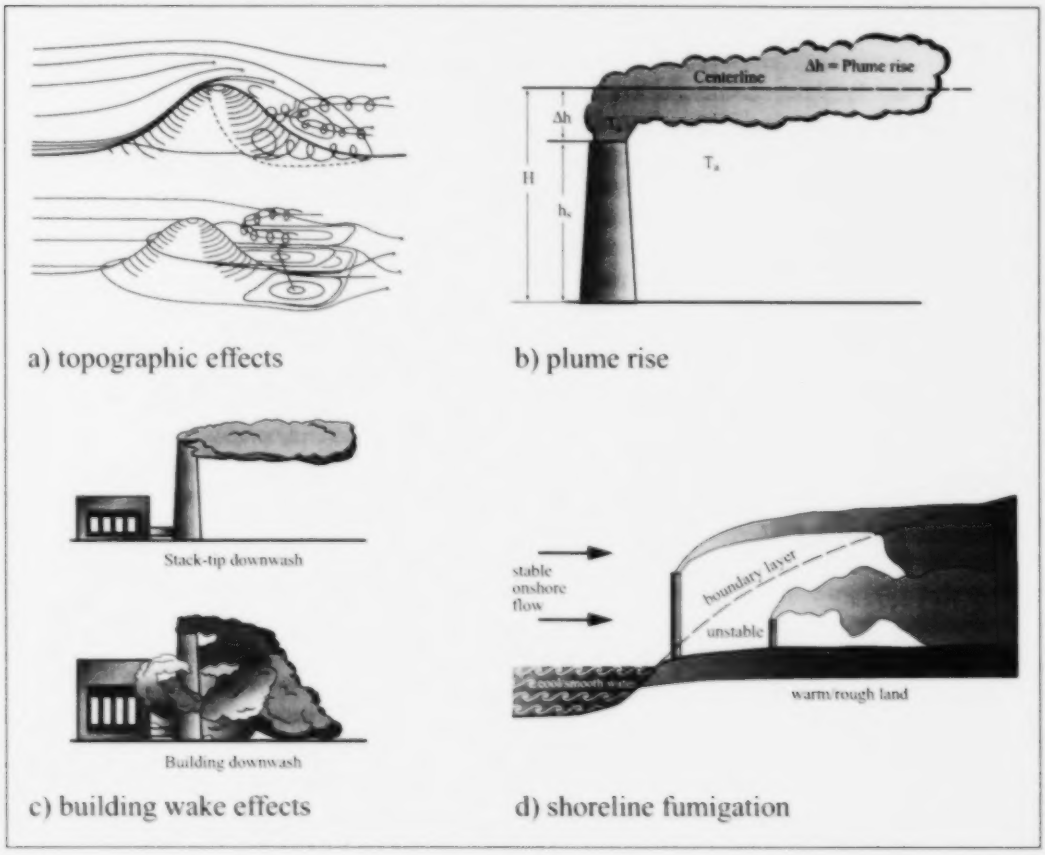
3.3.3.2 Effects of Topography

When a dispersing plume encounters a hill, it deflects up and over the hill, or around it, depending on the atmospheric conditions. The deflection is only partial, and as the plume passes over the hill, its height above ground is lower than it would be in the absence of the hill, which results in higher ground level concentrations. Simple Gaussian approaches do not explicitly represent dispersion around hills and need to be used with caution where significant hills are present between source and receptor.

This might be an important consideration for facilities located in more complex surroundings like Chalk River or unimportant for facilities located in relatively flat surroundings like Gentilly. The latest regulatory air quality models provide a better capability to handle complex surrounding conditions, which can be an important consideration for some facilities.

Where there is significant variation in surface roughness surrounding the source, the use of an average uniform roughness, may over or underestimate concentration at some locations, depending on the actual terrain. Considering the range in terrain roughness around Canadian nuclear facilities, the N288.1 Guideline recommends a lower value, which will be generally conservative with one exception. For receptors across water, vertical dispersion is over-estimated by assuming a roughness value for land surfaces.

Figure 3.2: Illustration of Atmospheric Plume Dispersion Processes

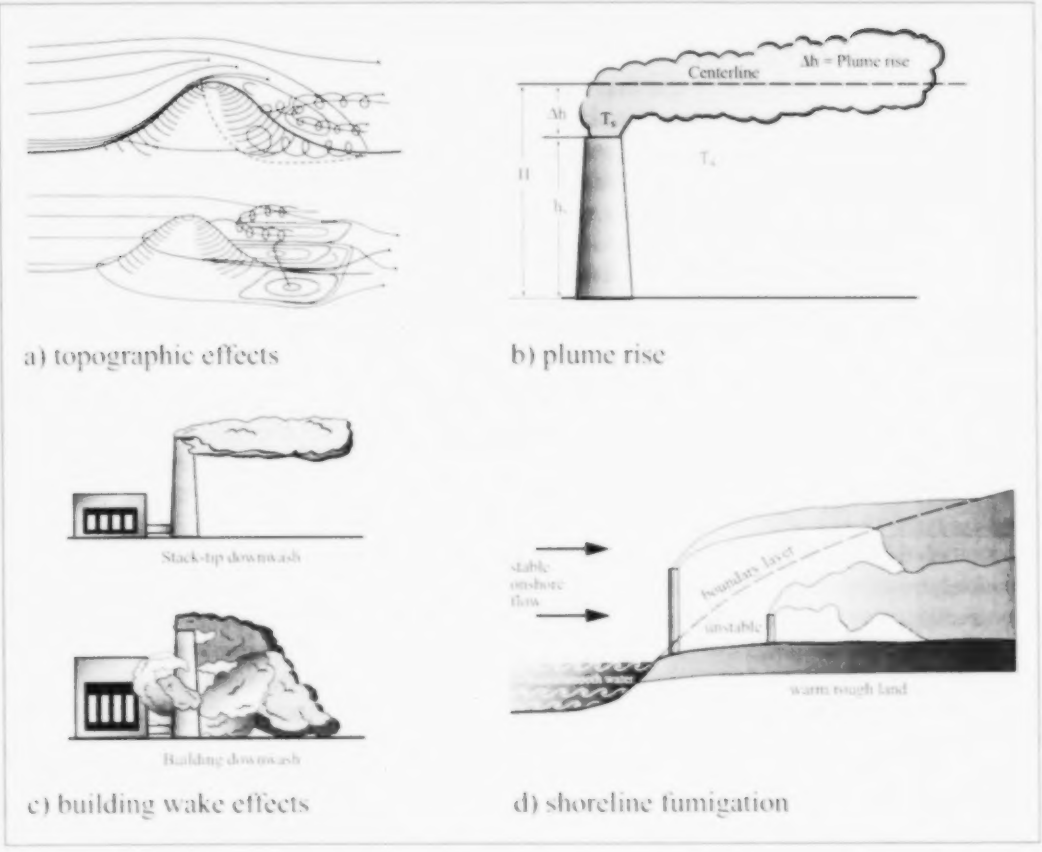


3.3.3.3 Plume Rise

Tritium plumes may rise following release from a stack, due to thermal buoyancy or momentum. As noted by Briggs (1971), effective release height H_{ik} can be adjusted up to account for plume rise, based on the dominant mechanism. For most reactor stacks, gas temperature is slightly above room temperature, and therefore thermal buoyancy is negligible in summer. It may be important in winter; however, an assumption of no buoyancy throughout the year is conservative in winter.

Plume rise stops when the driving force of plume heat or momentum is dissipated, usually within a few tens or hundreds of metres. The rise is countered by stack or building wake effects which draw the plume downward (Section 3.3.3.4). As noted in the N288.1 Guideline, calculations that ignore plume rise tend to overestimate ground-level concentration by less than 30% at most nuclear facilities. One exception is the Bruce incinerator where the stack gas temperature of 153°C results in significant plume rise.

Figure 3.2: Illustration of Atmospheric Plume Dispersion Processes



3.3.3.3 Plume Rise

Tritium plumes may rise following release from a stack, due to thermal buoyancy or momentum. As noted by Briggs (1971), effective release height H_s can be adjusted up to account for plume rise, based on the dominant mechanism. For most reactor stacks, gas temperature is slightly above room temperature, and therefore thermal buoyancy is negligible in summer. It may be important in winter; however, an assumption of no buoyancy throughout the year is conservative in winter.

Plume rise stops when the driving force of plume heat or momentum is dissipated, usually within a few tens or hundreds of metres. The rise is countered by stack or building wake effects which draw the plume downward (Section 3.3.3.4). As noted in the N288.1 Guideline, calculations that ignore plume rise tend to overestimate ground-level concentration by less than 30% at most nuclear facilities. One exception is the Bruce incinerator where the stack gas temperature of 153°C results in significant plume rise.

Condensation of gaseous HTO may occur at or very near the stack as hot gases cool and lose moisture, particularly in winter. Water collected from stack walls and roofs near the point of release may contain high concentrations of tritium. Runoff of such water may have a significant effect on local soil porewater and groundwater near the stack.

3.3.3.4 Stack and Building Wakes

Depending on the height and size of stack, the basic pattern of dispersion may be altered by aerodynamic effects of adjacent buildings and the stack itself. Turbulent zones (known as wakes) occur on the leeward sides of these structures as the wind flows around them. A tritium plume can, at least under some wind conditions, be drawn downward into the wakes, bringing the plume closer to the ground. This effect, known as downwash, can be significant for both short-term and long-term plume concentrations. If some of the tritium is present in the form of airborne particles or droplets, these particles will undergo gravitational settling which may also bring the plume downward somewhat. This will result in greater contaminant concentrations in the near field.

The model outlined in N288.1 includes downwash effects. It uses a simplified approach based on a method developed by Huber (1984), in which the effective release height (H_{ts}) is reduced by an amount that depends on the inside stack diameter and exit velocity, and the height of adjacent buildings, and the plume spread is enhanced by an amount that is calculated based on the cross-sectional area of adjacent buildings. Relevant stack and building parameters are shown in Table 3.5. If exit velocity exceeds 1.5 times wind speed, downwash at the tip of the stack will not occur. However, downwash in the lee of the building may still occur, depending on stack height and building dimensions. If the stack height adjusted for stack tip downwash exceeds 2.5 times the height of adjacent buildings, plume entrainment by buildings will not occur. Full entrainment may be reasonably assumed when stack-height is less than the height of adjacent buildings, which is typical of many Canadian reactor sites.

It is often not obvious how to choose the cross-sectional area, since releases from most nuclear sites are affected by several different buildings. Newer generation dispersion models have replaced this approach with a somewhat more sophisticated approach (the so-called PRIME algorithm).

Table 3.5: Stack and Building Parameters Relevant to Atmospheric Dispersion of Tritium from Canadian Reactor Sites. (from Hart, 2008).

Site	Stack Height (m) (h_s)	Adjacent Building Height (m) (h_b)	Stack* Inside Diameter (m) (D)	Stack* Exit Velocity m/s (W_o)	Adjacent Building Cross-section Area (m^2)
Pickering	41	43	1.27	12.9	1850
Darlington	59	70	2.79	19.3	2500
Gentilly-2	37	46	-	-	3000
Point Lepreau	50	-	-	-	3000
Bruce	-	-	2.32	16.2	1950
Bruce Incinerator	21	13.7	0.41	13.1	1350
CRL	46	0	-	-	0
CRL Mo-99	61	<<61	-	-	1500

* *reactor stacks generally have an inside diameter in the 1-3m range, and exit velocity in the 10-20 m/s range; parameters shown are for the highest flow stack when several are present at a facility*

** *at Bruce some stacks are lower than the containment building, while others are slightly higher; it is assumed that $h_s < h_b$ so that the plume is fully entrained.*

Notes:

$H_k = h_s - (\Delta h_d)_k - (\Delta h_{em})_k$ for a neutrally buoyant plume.

$(\Delta h_d)_k = 2\,(1.5 - w_o/u_k) \cdot D$

$(h')_k = h_s - (\Delta h_d)_k$

$(\Delta h_{em})_k = 1.5\, h_b - 0.6\,(h')_k$

Another factor to note is that some Canadian nuclear facilities release tritium to the atmosphere through a number of stacks or vents. Rather than modelling each stack individually, releases from a given facility are often approximated by combining them into one virtual source located at the centre of that facility. Prediction errors arising from this assumption are small (10% or less at PNGS). Also, the N288.1 modelling approach assumes a uniform continuous source emission rate and uses long-term average atmospheric conditions. It is not meant to be applicable if a facility releases more than about 1% of the annual DRL in a matter of a few hours or during adverse weather conditions (poor dispersion) (CSA 2008).

New generation models like AERMOD calculate dispersion on an hour-by-hour basis using hourly meteorological data rather than using statistically weighting to calculate long-term average concentrations. This allows for estimating potential exposures for various averaging times (down to 1-hour) under a full range of meteorological conditions and release rates. Such capability is important when looking at short-term high level exposures, but less important when considering long-term average exposures.

3.3.3.5 Shoreline Fumigation

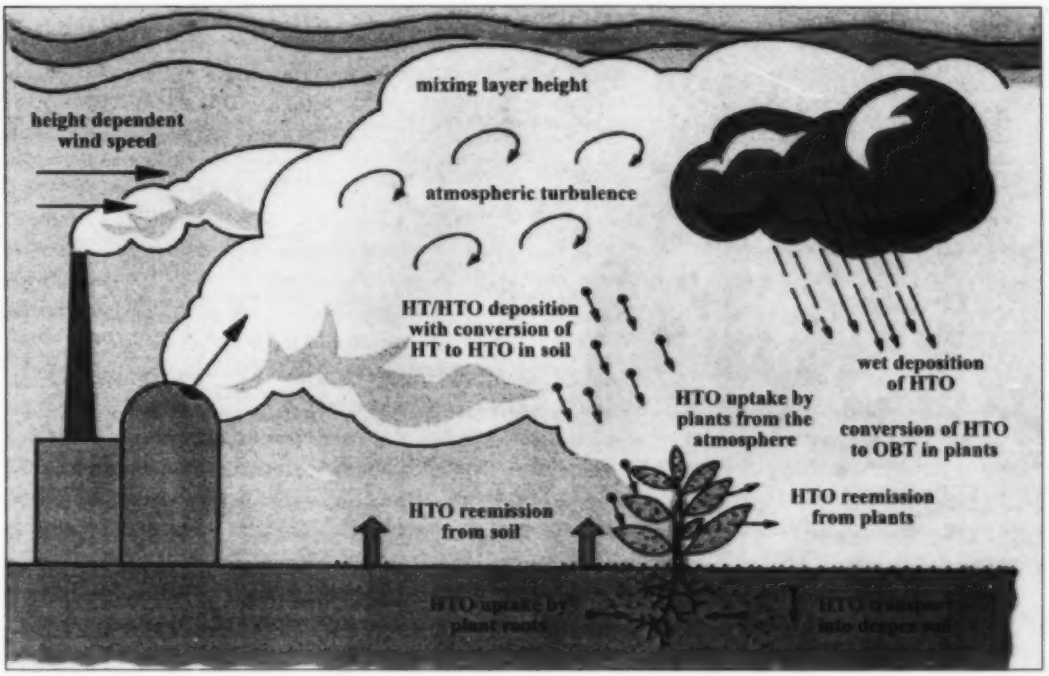
For stacks located at the shore of a large body of water and tall enough not to be affected by building aerodynamics, shoreline fumigation is another phenomenon that can alter the basic pattern of dispersion and draw the plume closer to the ground. This phenomenon can occur on low-wind, clear mornings when the wind flow is in an onshore direction. Early in the morning, the atmospheric condition is stable. Later in the morning, it remains stable over the water but starts to become unstable over the land, as the sun heats the earth's surface. At this time, the plume may at first be emitted into a stable environment with limited dispersion and limited plume rise. When it meets the developing unstable layer over the land, at the thermal inversion boundary layer (TIBL), the rate of dispersion increases rapidly, causing some of the plume to reach the ground sooner and at higher concentration than it otherwise would.

Shoreline fumigation could be significant for short-term exposure to a tritium plume released from a tall stack. While this process is not addressed by N288.1, shoreline fumigation is a relatively infrequent condition and most nuclear facilities have relatively short stacks. The stacks are generally 30 - 60m in actual height and effective release heights are even lower, particularly when adjacent building heights result in substantial plume entrainment. The effective release heights are generally below the height of the TIBL at the inland distance of Canadian nuclear sites. As such, shoreline fumigation is not expected to have a significant effect on long-term tritium concentrations.

3.3.4 Dry and Wet Deposition

Once released to the atmosphere, tritium may be deposited on vegetation, soil and other surfaces. Deposition may occur through various processes, as illustrated in Figure 3.3, which is taken from Galeriu *et al.* (2007). Tritium can either be taken up directly from the plume by vapour exchange with soil and plants (a process known as dry deposition), or can be scavenged from the plume by rain drops and deposited in rainfall (a process known as wet deposition or washout). Figure 3.3 also indicates that HTO may be re-emitted from soil or plants to atmosphere.

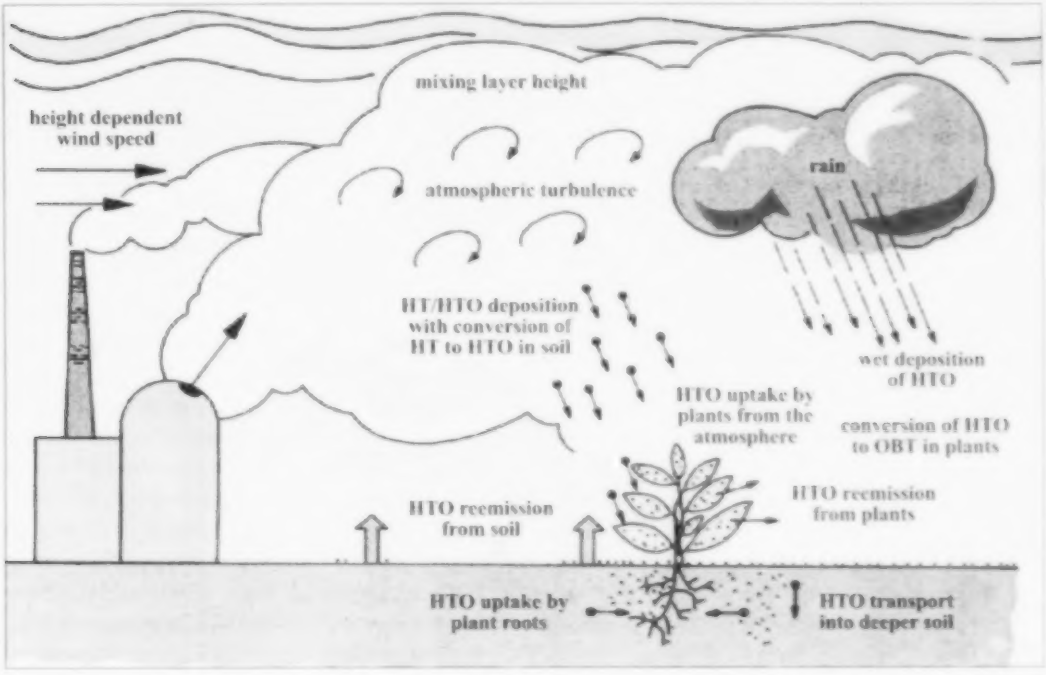
Figure 3.3: Environmental Tritium Processes (Galeriu, *et al.*, 2007)


mixing layer height
height dependent wind speed
atmospheric turbulence
HT/HTO deposition with conversion of HT to HTO in soil
wet deposition of HTO
HTO uptake by plants from the atmosphere
conversion of HTO to OBT in plants
HTO reemission from plants
HTO reemission from soil

An additional mechanism of tritium transfer from an air plume to soil involves condensation of tritiated water vapour onto the soil surface (i.e., formation of dew). Bunnenberg, *et al.*, (1986) found that, under certain meteorological conditions, there is a measurable exchange of tritiated water vapour between the atmosphere and soil due to this mechanism. These authors suggested that the condensation pathway is especially important for releases near ground level. In the case of an accidental release from a low-level source, under appropriate conditions, condensation could yield the greatest depositions of HTO. Generally, it is a less important mechanism as compared to vapour exchange and washout.

Tritium transfer from air to soil water, groundwater and surface water is discussed in detail in Section 4.0, as an aspect of tritium behaviour in the hydrological cycle. It is mentioned here in the context of depletion of the airborne plume. The N288.1 modelling approach conservatively ignores depletion of the airborne plume due to wet or dry deposition. However, the effect of depletion of the airborne concentration of tritium due to these processes has been evaluated by calculating concentrations with and without depletion for potential critical groups at various nuclear generating stations in Canada (Hart, 2008). Calculations were done for two potential critical groups at each site, one located close to the reactors and one farther away. Average values were adopted for the deposition parameters corresponding to the observed precipitation intensity. Results of the study indicate that modeling without depletion tends to overestimate the concentrations by around 10% near the source (1 to 2 km away), and 20 to 35% at longer distances (4 to 10 km).

Figure 3.3: Environmental Tritium Processes (Galeriu, *et al.*, 2007)


mixing layer height
height dependent wind speed
atmospheric turbulence
rain
HT/HTO deposition with conversion of HT to HTO in soil
wet deposition of HTO
HTO uptake by plants from the atmosphere
conversion of HTO to OBT in plants
HTO reemission from plants
HTO reemission from soil
HTO uptake by plant roots
HTO transport into deeper soil

An additional mechanism of tritium transfer from an air plume to soil involves condensation of tritiated water vapour onto the soil surface (i.e., formation of dew). Bunnenberg, *et al.*, (1986) found that, under certain meteorological conditions, there is a measurable exchange of tritiated water vapour between the atmosphere and soil due to this mechanism. These authors suggested that the condensation pathway is especially important for releases near ground level. In the case of an accidental release from a low-level source, under appropriate conditions, condensation could yield the greatest depositions of HTO. Generally, it is a less important mechanism as compared to vapour exchange and washout.

Tritium transfer from air to soil water, groundwater and surface water is discussed in detail in Section 4.0, as an aspect of tritium behaviour in the hydrological cycle. It is mentioned here in the context of depletion of the airborne plume. The N288.1 modelling approach conservatively ignores depletion of the airborne plume due to wet or dry deposition. However, the effect of depletion of the airborne concentration of tritium due to these processes has been evaluated by calculating concentrations with and without depletion for potential critical groups at various nuclear generating stations in Canada (Hart, 2008). Calculations were done for two potential critical groups at each site, one located close to the reactors and one farther away. Average values were adopted for the deposition parameters corresponding to the observed precipitation intensity. Results of the study indicate that modeling without depletion tends to overestimate the concentrations by around 10% near the source (1 to 2 km away), and 20 to 35% at longer distances (4 to 10 km).

3.3.5 Next Generation Dispersion Models

The US EPA (2003) conducted a comprehensive assessment of the differences between earlier dispersion models (i.e., ISC) with newer generation models such as AERMOD. Since the model often used to predict contaminant concentrations at Canadian nuclear facilities is similar to ISC, the findings of this assessment can be used to provide some general comments regarding the differences between ISC-like models and more advanced regulatory models like AERMOD. The US EPA (2003) found that relative to ISCST3, AERMOD currently contains new or improved algorithms for:

- Dispersion in both the convective and stable boundary layers;
- Dry and wet depositions;
- Plume rise and buoyancy;
- Plume penetration into elevated inversions;
- Treatment of elevated, near-surface, and surface level sources;
- Computation of vertical profiles of wind, turbulence, and temperature; and,
- 'Continuous' treatment of receptors on all types of terrain from the surface up to and above the plume height (i.e., simple, intermediate, and complex terrain).

For non-downwash settings in flat and simple terrain, AERMOD tends to predict maximum concentrations that are similar to ISC. Where building downwash is a significant factor in the air dispersion analysis, AERMOD again predicts maximum concentrations and maximum cavity concentrations that are very similar to ISC. More significant differences between the two models occur when dealing with dispersion around hills. AERMOD tends to produce much lower maximum concentrations than the screening technique in ISC. Overall, the US EPA (2003) concluded that AERMOD outperforms ISC (-ST3 and –PRIME).

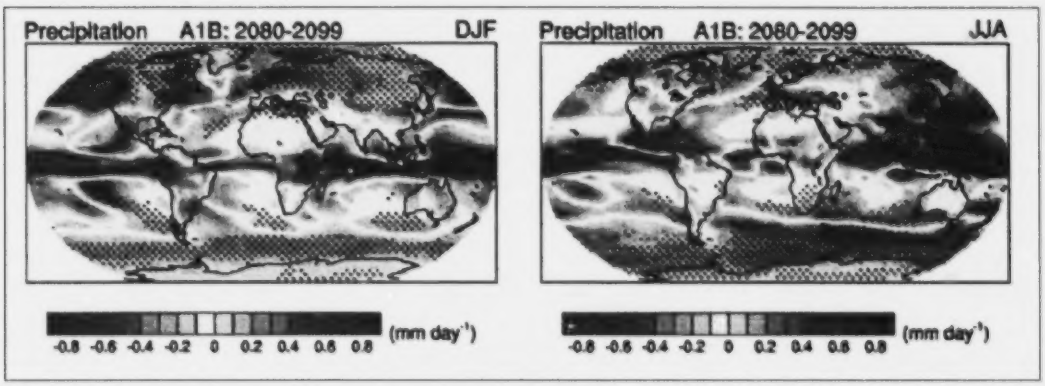
3.4 Impact of Climate Change on Tritium Dispersion

In recent years, there has been increased interest in understanding the influence of climate change on weather patterns, atmospheric chemistry processes and future air quality. Studies indicate that continued Green House Gas (GHG) emissions at or above current rates would cause further warming and induce many changes in the global climate system during the 21st century that would very likely be larger than those observed during the 20th century (IPCC, 2007, MacCracken, 2008). These factors have the potential to influence the atmospheric dispersion of radionuclides released from nuclear facilities.

There is now higher confidence in projected patterns of warming and other regional-scale features, including changes in wind patterns, precipitation and some aspects of extreme weather. Factors such as changes in precipitation, absolute humidity in the air and soil moisture content will be particularly important for tritium behaviour in the atmosphere since partitioning from air to surface water or soil water depend on these parameters. According to the IPCC (2007), future climate change projections using multi-model ensembles show increases in globally averaged mean water vapour, evaporation and precipitation. Of importance to Canada, the models suggest that precipitation will be

greater in the areas at high latitudes, with increases in annual precipitation exceeding 20 percent. In most of Eastern Canada and southern Ontario, projections show that compared with 1980-1999 averages the mean precipitation for 2080-2099 in winter (December to February) will increase (+0.2 ~ +0.4 mm/d), while mean precipitation in summer (June to August) will decrease at -0.1 mm/d, as shown by the predicted mean changes in precipitation presented in Figure 3.4 below.

Figure 3.4: Predicted Average Winter and Summer Precipitation for the Years 2080-2099 (IPCC, 2007)



Overall, precipitation may increase by about 4 percent and absolute humidity is also expected to slightly increase. Likewise, soil moisture content in southern and eastern Ontario may increase by as much as 5 percent. Assuming these predictions are true, and future annual average precipitation rates, humidity and soil moisture increase, it is expected that this change will affect tritium deposition and may slightly increase HTO deposition to soil and ground water. This could lead to an increase in uptake through these pathways. At the same time, increased deposition may decrease annual average airborne concentrations somewhat, resulting in a decrease in inhalation exposure.

An increased frequency of extreme weather events over the past few decades has been noted (Environment Canada, 1998). These include tropical storms and hurricanes at tropical and subtropical latitudes, and winter storms at temperate latitudes. While these are believed to be linked to climate forcing by GHG, waves of extremes may also represent natural climate cycles. Several studies have looked at the frequency and severity of weather across Canada and the United States. The Climatic Loads Task Force (Auld *et. al.*, undated) reports that the average annual precipitation increased approximately 12% between 1900 and 1998 in southern Canada. They also cite a number of U.S. studies that report an upward trend in the frequency of heavy and extreme precipitation events in many southern regions across Canada, including the Great Lakes basin. The results of climate change models suggest that further increases in extreme precipitation events in southern Canada will occur over the next several decades. In many locations, what was previously considered to be the 50 year storm is predicted to occur with a significantly shorter return period in future.

There remains a high level of uncertainty regarding the impact of climate change on the dispersion and behaviour of tritium in the atmosphere. One of the reasons is that climate change modeling currently does not reliably predict effects on short-term (day-to-day) weather variance, which plays a significant role in tritium dispersion. Also, the resolution of current climate change models is regional, which is beyond the more localized effects of sub-regional features (shorelines, topography, etc.) that surround individual facilities. Thus, more detailed work on climate change downscaling techniques is needed to fully understand the influence of climate change on the atmospheric dispersion of tritium at nuclear facilities.

4.0 DYNAMIC BEHAVIOUR OF TRITIUM IN THE HYDROLOGICAL CYCLE

This section outlines the partitioning of tritium from air to soil water and surface water, tritium migration from soil water to groundwater, and tritium behaviour in lake and river plumes. The transfers between air, soil water, surface water and groundwater are highlighted in Figure 4.1. For context, this figure also illustrates the full set of transfers between environmental compartments, relevant to human exposure and dose assessment.

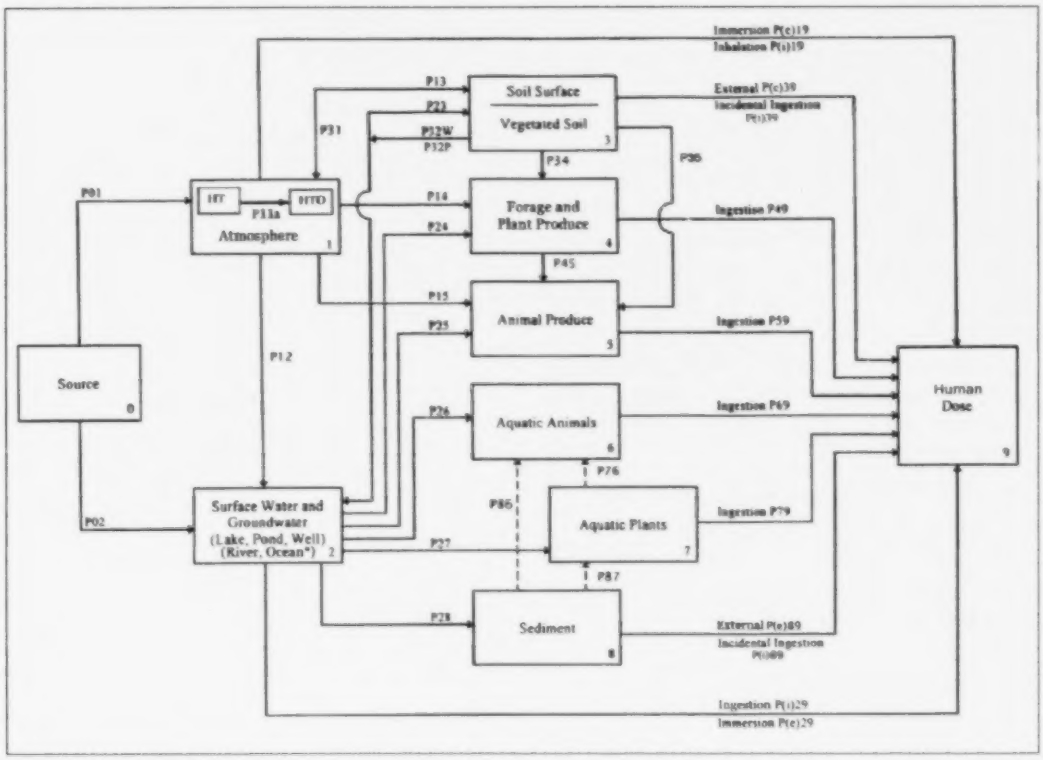
4.1 Partitioning from Air to Soil and Surface Water

As noted in Section 3.1.3, tritium may be transferred by vapour exchange and by precipitation between the air and the surface of the earth, i.e. to soil porewater and to surface water bodies. The vapour exchange is continuous, and is the dominant process for plumes near the ground (Bunnenberg *et al.*, 1986). This exchange is augmented by transfer of tritium in precipitation during rainfall and snowfall events. For snowfall, the transfer is completed when the snow melts (Davis, 1997). Light rainfall events are more effective than heavy ones in scavenging tritium from a local air plume (Bolin, 1961; Sejkora, 2006).

It is difficult, and generally unnecessary, to separate the transfer of tritium that occurs via precipitation, from that which occurs via direct water vapour exchange. Tritium concentrations in a body of surface water or in soil porewater respond to the recent history of tritium inputs and reflect a dynamic equilibrium between air and water. The relevant historical period depends on the residence time of water in the water body or the surface soil. Water residence times in surface soil are on the order of months. Those in deeper sub-soils can range from years to thousands of years. Residence times in ponds and lakes range from weeks to hundreds of years, depending on their volumes and discharge rates.

The transfers from air to soil water, or to small surface water bodies such as ponds, are best represented by specific activity models. These models apply when there is rapid isotopic exchange between environmental compartments, so that if one compartment is initially contaminated, the other compartment will tend to have the same tritium to hydrogen (T/H) ratio, except for any dilution effects. The T/H ratio is referred to as the specific activity of tritium.

Figure 4.1: Environmental Transfer Model



- **For ocean water, pathways P_{23}, P_{24}, P_{25} and $P(i)_{29}$ are not used.**

The following sections consider tritium transfer from air to surface soil water, and from air to surface water, separately, since there are substantial differences between these environmental compartments.

4.1.1 Partitioning from Air to Soil Water

The transfer of tritium as HTO in water vapour (or rain) to HTO in soil water is best represented by means of a specific activity model. This model assumes that soil water will have the same specific activity as air moisture, except for dilution due to uncontaminated precipitation. The model can be formulated as follows (Hart, 2008):

$$P_{13sp_{sw}\_HTO} = RF_{sw} / H_a \quad (m^3 \cdot L^{-1}) \qquad \text{Equation 4.1}$$

where: RF_{sw} = the ratio of HTO concentration in soil water to that in air moisture ($Bq \cdot L^{-1}$ soil water per $Bq \cdot L^{-1}$ air moisture)

H_a = atmospheric absolute humidity ($L \cdot m^{-3}$)

Empirical data for RF_{sw} are available from a number of reactor sites, including the Pickering NGS (Spencer and Vereecken-Sheehan, 1994) and the Chalk River Laboratories (CRL). The CRL data (Davis *et al.*, 2002) were contributed to the BIOMASS program, and appear in Appendix B of the final BIOMSS report (IAEA, 2003), along with Russian and French data sets in Appendices C and D. The various data sets (Tables 4.1 and 4.2) are consistent with a default value of 0.3 for RF_{sw}, representing dilution of soil water by uncontaminated precipitation, which falls when the atmospheric plume is not over the soil.

Table 4.1: Values of RF_{sw} Measured at Pickering NGS (Spencer and Vereecken, 1994)

Date	Soil Water[1] (Bq/mL)	Air Moisture[2] (Bq/mL)	Ratio (RF_{sw})
21 July	0.84	3.37	0.25
06 August	1.29	1.74	0.74*
10 September	2.42	7.51	0.32
24 September	1.82	7.93	0.23
07 October	2.2	9.51	0.23
22 October	1.33	4.8	0.28
06 November	1.66	5.16	0.32
Average (Mean)			**0.34**
Standard Deviation			**0.18**

[1] *From a soil depth of 0-7 cm.*

[2] *Collected by passive sampler.*

* *Outlier value included in statistical analysis.*

Table 4.2: Values of RF_{sw} Measured at Three Reactor Sites (IAEA, 2003)

Location on Site	Canada (CRL)[1]	Russia (RFNC)	France (CEA)
1	0.31	0.261	0.27
2	0.26	0.131	0.44
3	0.22	0.344	0.24
4	-	-	0.17
Average (Mean)	**0.26**	**0.25**	**0.28**
Standard Deviation	**0.045**	**0.11**	**0.11**

Notes:

The three sites are not significantly different (ANOVA, $p = 0.898$). For pooled data, $M = 0.265$, $SD = 0.087$.

[1] *For a soil depth of 0.10 cm, average values, passive sampler data for air (Davis et al., 2002).*

Table 4.3: Regional Absolute Humidity and Tritium Transfer from Air to Soil Porewater

Parameter Name	Darlington	Pickering	Bruce	CRL	G-2	Point Lepreau
Ha ($L \cdot m^{-3}$) for transfer to soil pore water	0.0069	0.0069	0.0066	0.005	0.0047	0.0054
RF_{sw} (unitless)	0.3	0.3	0.3	0.3	0.3	0.3
P_{13spw} ($m^3 \cdot L^{-1}$)	43.5	43.5	45.5	60.0	63.8	55.6

Absolute humidity from Environment Canada's Atmospheric Environment Service 1971-2000; RF_{sw} based on data in Tables 4.1. and 4.2

The absolute humidity, H_a, should be an annual average value in order to represent annual average soil water for use in annual dose calculations. Site-specific data should be used if available, or default regional values can be used (Table 4.3). Based on regional Canadian data, default values of P_{13spw} for tritium are shown in Table 4.3.

In the absence of a point source, it is reasonable to expect that the tritium concentration in soil water will be equal to that in air moisture, the underlying premise of the model. Pre-operational baseline data for the Cernavoda site in Romania (Paunescu *et al.*, 1999) indicates that tritium in soil water was similar to that in rain water (approximately 4 Bq/L). The baseline data for a site on Mt. Hakkoda in Japan (Sasaki *et al.*, 2000) indicates that the tritium concentration in water vapour is slightly higher than that in precipitation (1.1 vs 1.0 Bq/L). Thus, the model premise may be slightly conservative.

Davis *et al.* (1995) and Noguchi *et al.* (2001) reported on short-term variation in the HTO transfer soil water and to samplers containing water following a chronic HT release experiment. Some of the HT released was converted to HTO, as discussed in Section 3.2.1. The HTO in air moisture and soil or sampler water was measured over a 12 day period following HT release. It was found that HTO in soil water was usually slightly higher than that in air moisture, reflecting the soil origin of HTO by conversion of HT, and that rainfall events served to dilute the soil water by a factor of two or more, as compared to rain-free periods. The samplers indicated that there was a natural diurnal cycle in the rate of HTO transfer to water. The exchange velocities ranged from a low of 10^{-8} m/s at night to a high of 10^{-7} m/s during the day. Correlations between exchange velocities and meteorological conditions showed a strong correlation with wind speed, and weaker correlations with solar radiation and air temperature.

Under HTO release conditions, higher rates of HTO transfer to soil water are reported. Davis *et al.* (1997) reported a dry deposition velocity of 1.6×10^{-3} m/s to snow over an 18 day period of HTO release in winter, while Barry (1964) reported 3.0×10^{-3} m/s under similar conditions (these depositions impact soil water in the spring). Under summer conditions, dry deposition velocities of 4.0×10^{-3} m/s (Gulden *et al.*, 1990) and 20×10^{-3} (Bunnenberg *et al.*, 1992) have been reported.

Neither diurnal nor seasonal variations in rates of exchange or deposition affect the expectation of similar specific activity in air moisture and soil water. However, these rates do influence the time needed for an air moisture - soil water system to achieve the expected equilibrium after a change in the system, such as a shift in the air plume to a new compass sector. In a point source situation, the tritium in soil water will be constantly tracking the changes in overhead air moisture, with a lag time, and will rarely be precisely at equilibrium.

Given an interest in annual average exposure and dose, it is not necessary for tritium partitioning models to track the diurnal cycle, or other fine scale variations in partitioning. It is important for partitioning estimates to be accurate on an average annual basis.

The residence time of water or HTO in the shallow soil compartment may be approximated as the shallow soil depth divided by the rate of water infiltration through the shallow soil zone. In cultivated soils, the top 20-30 cm are mixed at least annually, while in other soils natural bioturbation results in mixing to approximately 20 cm. Given infiltration rates in the range of 0.2-0.4 m/a (Baron, *et al.*, 1997; Lin and Wei, 2005; Hart, 2008) soil residence times may be expected to range from 0.5 to 1.5 years. The corresponding hydological half-lives are 0.35 to 1.05 years. Based on this range of half-lives, once tritium input to the soil stops, the tritium concentration in shallow soil water can be expected to fall below 10% of the concentration when input ceased, within 2 to 4 years, or 5% within 2 to 5 years.

4.1.2 Partitioning from Air to Surface Water

The transfer of tritium as HTO in water vapour (or rain) to HTO in a pond has been represented as a transfer to precipitation, as a conservative assumption, since pond hydrology tends to be site-specific, and since pond data are generally lacking. Any surface water inflow or shallow groundwater inflow originating further away from the atmospheric point source is likely to carry less tritium contamination than precipitation, and will therefore provide dilution. However, it is conservative to assume no such dilution.

In theory, Equation 4.1 could be used to estimate tritium transfer from air to pond, with a reduction factor (RF) to account for the dilution by uncontaminated precipitation which falls when the local plume is not over the pond or its catchment area. However, since ponds are quite variable in size and catchment area, it would be difficult to develop a generic reduction factor, even if measurements on some ponds were available.

Based on the conservative assumption that pond water is like precipitation, many reactor facilities have routinely monitored tritium (as HTO) in air and precipitation at their site boundaries, and have used the data to develop a conservative but generic air to pond transfer factor (P_{12p}). LaMarre (2000) suggested that $P_{12p} = 100$ m^3/L, based on data for the Pickering NGS, and this value was cited by Hart (2008) in developing guidance for DRL calculation at nuclear facilities. The relevant data for three facilities are presented in Table 4.4, including more recent data for Ontario Power Generation (Borromeo, 2007) and Bruce Power (McDougall, 2007).

It is evident from the Pickering data in Table 4.4 that an updated average P_{12p} value for the site would be closer to 80 m^3/L. Average values for the Darlington and Bruce sites would be 75 and 71 m^3/L, respectively. However, it is reasonable to retain 100 m^3/L as a somewhat conservative generic transfer factor for ponds.

For larger lakes, current-driven water movements, and water depth in the off-shore region, limit the influence of airborne tritium on the water column as it passes beneath an atmospheric plume. Hart (2008) presents a bounding calculation showing that dose per unit release to air is increased by less than 1% if air to lake water transfer is included as a pathway in the dose estimation. Thus, air to large lake transfer is usually not considered.

Tritium behaviour in aquatic plumes following a direct release to a large lake or river is the most important variable to understand in the context of dose per unit release to water. This aspect of tritium behaviour is discussed in detail in Section 4.3.

Table 4.4: Precipitation and Air Monitoring Data Relevant to Air to Pond Transfer Factor

Year	Darlington Precipitation Bq/L	Air Bq/m^3	P_{12p} m^3/L	Pickering Precipitation Bq/L	Air Bq/m^3	P_{12p} m^3/L	Bruce Precipitation Bq/L	Air Bq/m^3	P_{12p} m^3/L
1988	31	0.7	44.29	2900	27	107.41	440	6.6	66.67
1989	140	1.5	93.33	2100	22	95.45	484	7.8	62.05
1990	50	0.9	55.56	1300	17	76.47	295	4.9	60.20
1991	140	1.2	116.67	1350	14	96.43	235	4	58.75
1992	39	0.7	55.71	1150	12	95.83	279	3.9	71.54
1993	40	0.7	57.14	650	8	81.25	354	5.1	69.41
1994	62	0.9	68.89	700	6	116.67	221	3.48	63.51
1995	70	0.9	77.78	750	7	107.14	184	2.81	65.48
1996	48	0.6	80.00	450	5	90.00	184	2.85	64.56
1997	68	0.6	113.33	700	6	116.67	140	2.3	60.87
1998	62	0.65	95.38	600	6	100.00	170	1.6	106.25
1999	80	0.65	123.08	550	5	110.00	149.6	2.20	68.01
2000	76	0.6	126.67	450	4	112.50	108	1.37	78.83
2001	76	1.2	63.33	300	9	33.33	106	1.6	66.25
2002	60	0.8	75.00	300	8	37.50	111	2.2	50.45
2003	28	0.6	46.67	250	6	41.67	112	1.67	67.07
2004	40	0.8	50.00	300	7	42.86	110.2	1.74	63.33
2005	26	0.7	37.14	250	8	31.25	225.6	1.9	118.74
2006	28	0.7	40.00	250	7	35.71	190	2.18	87.16
	Mean		74.74			80.43			71.01
	SD		29.10			32.12			16.62
	79th percentile		99.33	67th percentile		100.43	93rd percentile		101.29

LaMarre (2000) value of P_{12p} = 100 m^3/L is based on Pickering data from 1988-2000
Tabulated data from Borromeo (2007) and McDougall (2007)

It is evident from the Pickering data in Table 4.4 that an updated average P_{12p} value for the site would be closer to 80 m^3/L. Average values for the Darlington and Bruce sites would be 75 and 71 m^3/L, respectively. However, it is reasonable to retain 100 m^3/L as a somewhat conservative generic transfer factor for ponds.

For larger lakes, current-driven water movements, and water depth in the off-shore region, limit the influence of airborne tritium on the water column as it passes beneath an atmospheric plume. Hart (2008) presents a bounding calculation showing that dose per unit release to air is increased by less than 1% if air to lake water transfer is included as a pathway in the dose estimation. Thus, air to large lake transfer is usually not considered.

Tritium behaviour in aquatic plumes following a direct release to a large lake or river is the most important variable to understand in the context of dose per unit release to water. This aspect of tritium behaviour is discussed in detail in Section 4.3.

Table 4.4: Precipitation and Air Monitoring Data Relevant to Air to Pond Transfer Factor

Year	Darlington Precipitation Bq/L	Air Bq/m³	P_{12p} m³/L	Pickering Precipitation Bq/L	Air Bq/m³	P_{12p} m³/L	Bruce Precipitation Bq/L	Air Bq/m³	P_{12p} m³/L
1988	31	0.7	44.29	2900	27	107.41	440	6.6	66.67
1989	140	1.5	93.33	2100	22	95.45	484	7.8	62.05
1990	50	0.9	55.56	1300	17	76.47	295	4.9	60.20
1991	140	1.2	116.67	1350	14	96.43	235	4	58.75
1992	39	0.7	55.71	1150	12	95.83	279	3.9	71.54
1993	40	0.7	57.14	650	8	81.25	354	5.1	69.41
1994	62	0.9	68.89	700	6	116.67	221	3.48	63.51
1995	70	0.9	77.78	750	7	107.14	184	2.81	65.48
1996	48	0.6	80.00	450	5	90.00	184	2.85	64.56
1997	68	0.6	113.33	700	6	116.67	140	2.3	60.87
1998	62	0.65	95.38	600	6	100.00	170	1.6	106.25
1999	80	0.65	123.08	550	5	110.00	149.6	2.20	68.01
2000	76	0.6	126.67	450	4	112.50	108	1.37	78.83
2001	76	1.2	63.33	300	9	33.33	106	1.6	66.25
2002	60	0.8	75.00	300	8	37.50	111	2.2	50.45
2003	28	0.6	46.67	250	6	41.67	112	1.67	67.07
2004	40	0.8	50.00	300	7	42.86	110.2	1.74	63.33
2005	26	0.7	37.14	250	8	31.25	225.6	1.9	118.74
2006	28	0.7	40.00	250	7	35.71	190	2.18	87.16
	Mean		74.74			80.43			71.01
	SD		29.10			32.12			16.62
	79th percentile		99.33	67th percentile		100.43	93rd percentile		101.29

LaMarre (2000) value of P_{12p} = 100 m^3/L is based on Pickering data from 1988-2000
Tabulated data from Borromeo (2007) and McDougall (2007)

4.2 Tritium Transport from Soil Water to Ground Water

The transfer of tritium as HTO in soil water to the groundwater aquifer at any depth level is essentially a process of vertical downward infiltration. This is a simplification of the process because there is also horizontal flow in the aquifer, if a gradient exists, as well as upward flow where the aquifer intersects a surface watercourse, i.e. a discharge zone (Figure 4.2). Both longitudinal and lateral dispersion occurs along the horizontal flow path, so the plume arising from the source area becomes wider, but centre-line concentrations are reduced along the flow path. Tritium concentrations are also reduced by decay along the flow path.

Horizontal flow is most important for a receptor (i.e. well) in the downgradient direction from an atmospheric source, because the horizontal flow from upgradient may contain higher tritium concentrations than those arising from the air directly above the receptor. In other situations the transfer from soil water to groundwater may be conservatively represented as a vertical downward infiltration.

Hart (2008) assumed a simple vertical downward transport in defining P_{3spw2w}, the transfer of radionuclides from soil water to a groundwater well, as follows:

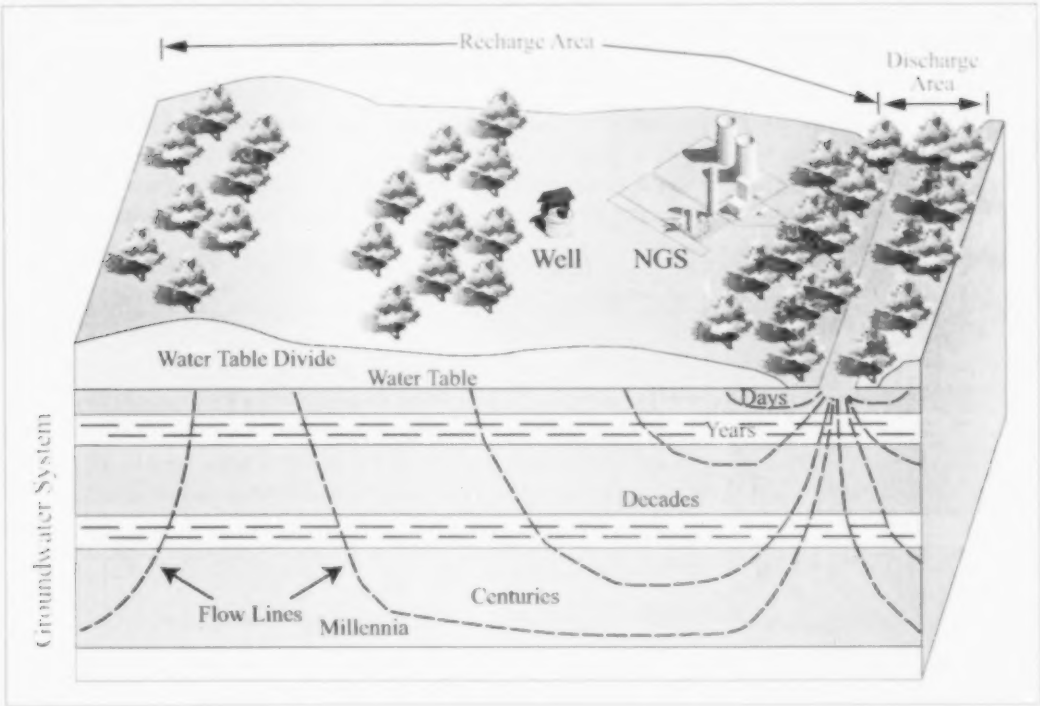
$$P_{3spw2w} = \exp(-\lambda_r \bullet T_r)$$
$$= \exp[-\lambda_r \bullet Z_{TOS}\,(n+K_d \bullet \rho_b) / q_{gw\,infil}] \quad (L \bullet L^{-1}) \qquad \text{Equation 4.2}$$

where:

T_r = travel time from surface soil to well screen (s)
Z_{TOS} = the soil depth to the top of the well screen (m)
n = effective porosity (unitless)
K_d = the partition coefficient of the radionuclide in the soil ($m^3 \bullet kg^{-1}$)
ρ_b = the soil bulk density ($kg \bullet m^{-3}$)
$q_{gw\,infil}$ = the infiltration rate (m/s)
λ_r = the radioactive decay rate (s^{-1})

In the case of tritium, the K_d is zero, since water does not adsorb to soil, and the term $K_d \bullet \rho_b$ drops from the equation.

Figure 4.2: Schematic of a Groundwater Flow System.

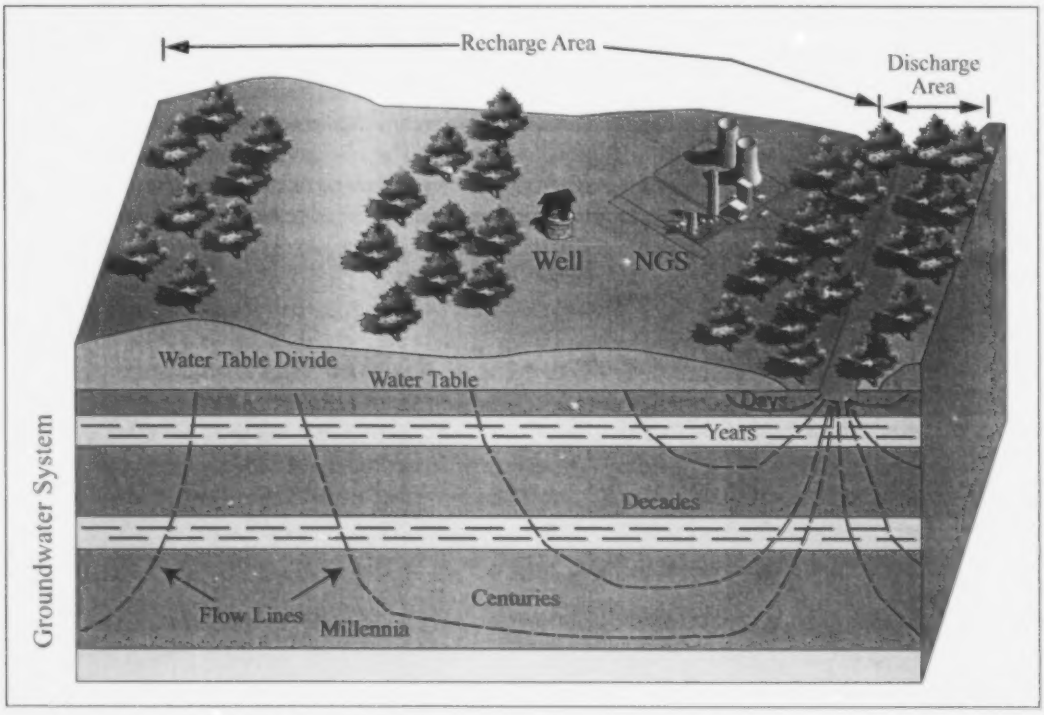


Site-specific parameter values are recommended, but may not always be available. Therefore default values have been defined. For $q_{in\ soil}$ a default value of 4.75×10^{-3} m • s^{-1} (0.15 m • a^{-1}) can be used, based on data from Jensen *et al.* (1995) at the Bruce Power site. For a deep well, the infiltration rate may be less than the infiltration to shallow soil, because some infiltration is usually lost as shallow horizontal flow and or evapotranspiration.

The effective porosity (n) represents an average for the entire pathway between the ground surface and the intake zone of the well. The soil moisture content can be considered an effective porosity in the unsaturated zone above the water table; thus, a value of 0.2 would be appropriate. The porosity of most unconsolidated porous media below the water table is in the range of 0.3 to 0.5. The porosity of fractured rock is lower, at about 0.2 for sedimentary rock and 0.1 for igneous rock. Overall, an effective porosity of 0.2 may be used if site-specific data are not available.

While the total depth of a given well is usually known, the depth to the well screen is not always available. For this reason, an assumption can be made that the intake zone represents the lower 20 percent of the total depth of the well. This assumption is reasonable because the well screen is usually located in the most permeable zone at depth, and it is desirable to minimize the length of the well screen for cost considerations. Wells will generally have a casing that extends from the top of the well screen to the surface, and that zone would not be permeable and would not allow water to enter the well.

Figure 4.2: Schematic of a Groundwater Flow System.



Site-specific parameter values are recommended, but may not always be available. Therefore default values have been defined. For $q_{gw\ infil}$ a default value of 4.75×10^{-9} m • s^{-1} (0.15 m • a^{-1}) can be used, based on data from Jensen *et al.* (1995) at the Bruce Power site. For a deep well, the infiltration rate may be less than the infiltration to shallow soil, because some infiltration is usually lost as shallow horizontal flow and/or evapotranspiration.

The effective porosity (n) represents an average for the entire pathway between the ground surface and the intake zone of the well. The soil moisture content can be considered an effective porosity in the unsaturated zone above the water table; thus, a value of 0.2 would be appropriate. The porosity of most unconsolidated porous media below the water table is in the range of 0.3 to 0.5. The porosity of fractured rock is lower, at about 0.2 for sedimentary rock and 0.1 for igneous rock. Overall, an effective porosity of 0.2 may be used if site-specific data are not available.

While the total depth of a given well is usually known, the depth to the well screen is not always available. For this reason, an assumption can be made that the intake zone represents the lower 20 percent of the total depth of the well. This assumption is reasonable because the well screen is usually located in the most permeable zone at depth, and it is desirable to minimize the length of the well screen for cost considerations. Wells will generally have a casing that extends from the top of the well screen to the surface, and that zone would not be permeable and would not allow water to enter the well.

If the depth to the top of the screen is known it can be used as the effective depth. As a default, however, the effective depth of the well intake zone is 80% of the total depth of the well, or $Z_{TOS} = 0.8 \bullet Z_{well}$. This assumption implies that only the top of the well intake zone supplies water to the well. In reality, the water in the well is derived from the entire well intake zone, with younger water at the top and older at the bottom. Under a state of constant loading from an atmospheric release, older water would be less contaminated due to a longer time available for decay of constituent radionuclides. The assumption that water is only from the top of the intake zone, i.e., the youngest possible water, provides an added level of conservatism in the well model.

The model does not allow for any leakage for tritiated soil water down the well casing. If such leakage happens, water at the well screen will be younger than represented by the model, and therefore more contaminated than predicted because there will have been less time for decay than expected.

Other potential sources of error in modeling tritium concentrations in wells include the assumptions about the infiltration rate, screen depth, and effective porosity of the subsurface soil. Groundwater travel time is inversely proportional to hydraulic conductivity, which is proportional to the square of grain size (Freeze and Cherry, 1979). Therefore, substrate related error in the groundwater model is likely to follow a lognormal distribution. A GSD of 2 is sufficient to encompass the range of grain size variability in sand, and a similar distribution of error in groundwater predictions is expected.

Background levels of tritium (as HTO) in groundwater can be significant, depending on depth, because tritium inputs from atmospheric deposition have been higher in the past. Tritium levels in global air and precipitation began to increase around 1950 as a result of nuclear weapons testing and peaked in 1963 with the Nuclear Test Ban treaty between the United States and the Soviet Union. They have been decreasing since that that time. Tritium levels in Ottawa precipitation peaked at approximately 600 Bq/L in 1963 (Mutch and Mahony, 2008).

Depth profiles of tritium in groundwater usually show a peak at a depth level corresponding to 1963. Retrospective studies in Japan (Miyamoto *et al.*, 1995) show a tritium peak in precipitation at approximately 100 Bq/L in 1963, and a corresponding peak in shallow groundwater (top of saturated zone) at approximately 60 Bq/L in 1964. The reduced concentration in groundwater in this zone was attributed mainly to dilution by discharge of deeper uncontaminated groundwater into the top layer of the aquifer. The residence time in the top layer was estimated at 3 years, which is insufficient for appreciable decay of tritium.

A groundwater study in China (Lin and Wei, 2005) shows tritium in a 1988 core profile peaking at 66 Bq/L at a depth of 6-7m, representing the fallout signature, and a 1997 core profile at the same location peaking at 27 Bq/L at a depth of 10m. The difference in concentration is partly due to decay and partly due to longitudinal dispersion during vertical migration (Figure 2.3). The rate of vertical migration was estimated at 0.25 to 0.30 m/a.

A similar study in France (Baran *et al*, 1997) shows tritium in three 2002 core profiles peaking at a depth of 9-10m, representing the fallout signature. The average infiltration rate over the 1963-2002 period was estimated at 0.20 to 0.21 m/a based on the peak centre of mass.

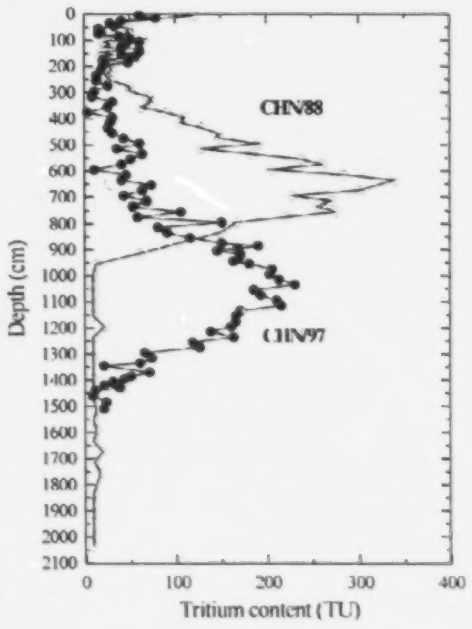


Figure 4.3: Changes Over Time in the Depth Profile of Tritium in Groundwater (from Lin and Wei, 2006).
(Note: 100 TU = 12 Bq/L; 1988 results are adjusted for decay to 1997 values)

4.3 Tritium Behaviour in Lake and River Plumes

Tritium as HTO is released from Canadian nuclear facilities via cooling water to large lake or river receiving environments. The Point Lepreau facility in New Brunswick releases to the Bay of Fundy, which can be Modelled as a large lake, with appropriate parameter adjustment. Gaussian type models for radionuclides released to large lakes and rivers are provided by the NCRP (1996) and have been widely used for estimating average annual concentrations of tritium in aquatic plumes arising from nuclear facilities.

The large lake model represents advection and dispersion processes driven by along-shore currents, as well as radionuclide losses due to radioactive decay and sedimentation. Since water travel times to receptor locations, i.e. drinking water intakes, are on the order of days, losses from radioactive decay are trivial. Tritium losses from sedimentation are zero. An initial dilution parameter allows for diffuser effects. The model as presented by Hart (2008) includes a factor for frequency of current direction towards the receptor location, so the concentration result at this location represents an annual average value. Other factors affecting the concentration result include water depth, current velocity and a lateral dispersion parameter.

Based on model calibration to temperature and/or tritium data, Hart (2008) provides default parameter values for Canadian facilities. Using these parameter values, the predicted tritium concentration increments at water supply plants (WSP) in the vicinity of the Pickering NGS agree reasonably well with background corrected measured values at the WSPs (Table 4.5). There is better agreement for farfield locations, where the plume is well dispersed, than for nearfield locations. Since the model predicts a plume centerline concentration, water intakes drawing from well off the centerline will experience lower than predicted concentrations, particularly in the nearfield.

The background level of tritium in the Great Lakes includes a component from fallout, which is declining, and a natural background component. The fallout component was estimated by Klukas (1999) to be 1.12 Bq/L in Lake Ontario in 2006. The natural background was estimated at 0.585 Bq/L. The combined background at present is approximately 1.70 Bq/L, unrelated to nuclear power stations.

There is a circulating background of HTO in Lake Ontario due to historical power station emissions, some of which are still circulating in the lake. The Pickering component of this can be estimated as follows, assuming historical releases similar to present-day releases:

$$\frac{dC}{dt} = \frac{W_e}{V} - \frac{C \bullet Q_{out}}{V} - C \bullet \lambda_r \qquad \text{Equation 4.3}$$

where:
- C = whole-lake concentration of HTO (Bq/L)
- W_e = loading of HTO from effluent (Bq/s)
- V = whole lake volume (m^3)
- Q_{out} = volumetric outflow rate (m^3/s)
- λ_r = radioactive decay constant (1/s)

Integrating this equation over time, the steady-state concentration (C) is as follows:

$$C = W_e / (Q_{out} + V \lambda_r) \qquad \text{Equation 4.4}$$

Based on the 2006 loading (W_e) of 1.05 x 10^7 Bq/s (Borromeo, 2007), outflow of 7,198 m^3/s (Environment Canada, 1983), and volume of 1.64 x 10^{12} m^3 (ILEC, 1999), the circulating background attributable to Pickering is estimated at 1.03 Bq/L. Another 0.6 Bq/L would be attributable to the Darlington station, based on 2006 releases. Levels measured at six far-field WSPs average 3.5 Bq/L, suggesting a circulating background of 1.8 Bq/L from power station sources, after correction for natural background and fallout. The difference between 1.6 and 1.8 Bq/L may reflect upstream inputs.

The river model from NCRP (1996) represents advection and dispersion processes driven by down-river currents, as well as radionuclide losses due to radioactive decay and sedimentation. Again, the loss terms are negligible within the travel times of interest. Default parameter values are provided by Hart (2008) for two reactor sites on rivers. Based on these parameters for the CRL site, a predicted tritium increment of 2.6 Bq/L at the Pembroke water intake 28 km downstream may be compared to a measured increment of 3.8 Bq/L (De Waele, 2006). A background of 3 Bq/L was measured upstream of the site.

Small variations exist in the background levels of tritium in lakes and rivers today, based on factors such as latitude and proximity to the sea. Radwan *et al.* (2001) reported background levels for inland rivers and tributaries in Poland at 1.5 to 1.9 Bq/L over the 1994-99 period. Lakes ranged from 1.4 to 1.8 Bq/L, while rivers near the Baltic Sea ranged from 0.7 to 1.3 Bq/L. The concentrations in inland waters were similar to those in precipitation, which ranged from 1.3 to 2.0 Bq/L over the same period. Miyamoto *et al.* (1995) reported tritium levels in Japanese rivers at 2.0 Bq/L in 1985, similar to average levels in precipitation.

Table 4.5: Simple Model of Aquatic Dilution with Distance (Local Plume) Superimposed on Circulating Background for Lake Ontario

	HTO Concentration (Bq/L)		Distance		Local Model	Total Model**	
Drinking Water Supply Plant	Measured Average	Average minus Background*	km	Direction	HTO increment	HTO increment	Adult Dose from Water Ingestion+
					Bq/L	Bq/L	µSv/a
Ajax	6.10	4.40	5	E	9.02	10.04	0.103
Whitby	6.37	4.67	12	E	3.99	5.02	0.051
Oshawa	7.14	5.45	19	E	2.48	3.51	0.036
Scarborough(Horgan)	5.11	3.42	11	W	3.75	4.78	0.049
Toronto (Harris)	5.15	3.45	22	W	1.82	2.84	0.029

* *Natural + fallout background = 1.7 Bq/L in 2006 (release = 1.05 x 10^7 Bq/s)*

** *Total model includes the local plume (centreline) plus circulating background (1.03 Bq/L)*

\+ *Dose based on average adult water consumption (511 L/a)*

5.0 PREDICTED AND OBSERVED ENVIRONMENTAL BEHAVIOUR OF TRITIUM AT RELEASE SITES

This section compares model predicted and observed concentrations of tritium in air, soil water, surface water and groundwater near selected tritium-releasing facilities. The facilities considered include tritium light manufacturers and nuclear reactor facilities. Environmental data sets that were used in developing predictive models (as discussed in Section 4.0) are not considered here, since data used to develop a model cannot be used to test its performance.

5.1 Tritium Light Manufacturers

Tritium light facilities include SRB Technologies Ltd. in Pembroke, Ontario, and Shield Source Inc. (SSI) in Peterborough, Ontario. Readily available data pertinent to annual average dispersion and environmental partitioning of tritium at these facilities were considered. An annual timeframe is appropriate given an interest in annual doses received by people, considering similar tritium residence times in soil, and considering that travel times to the point of groundwater use are usually much larger.

5.1.1 SRB Technologies Limited

Modelling and monitoring investigations of the SRB Technologies site were recently undertaken by EcoMetrix (2008). These investigations included modelling and measurement of tritium (HTO) concentrations in air, soil water and groundwater. The facility also releases HT; however, this is primarily of interest due to the small portion that converts to HTO. The HT itself has a much small dose coefficient and does not transfer to the human food chain.

Tritium concentrations in air were modelled using the IMPACT code, which contains a steady-state sector-averaged Gaussian plume model, for calculating derived release limits. The model used 2006 annual emissions (74,000 GBq/a[1]) and 2006 meteorological data. As a test of the air model, the predicted ground-level concentrations of tritium for 2006 were compared to annual average measured concentrations for the same year at 31 locations around the site (Figure 5.1), at distances ranging from 250 m to 2 km. The air monitoring was conducted by Atomic Energy of Canada Limited (AECL) using a passive sampling system.

The modelled vs. measured air concentrations are tabulated in Table 5.1. Their relationship is illustrated in Figure 5.2. The air model explained 85% of the measured spatial variation in tritium concentration ($R^2 = 0.85$). The model as implemented here does not represent plume rise due to temperature gradient, which may be expected to slightly reduce average modelled concentrations at ground level in the near field. As implemented, the model predictions were 83% greater than measured concentrations, on average. Approximately 60% of model

[1] Includes HTO emissions, plus 2% of HT emissions (0.02 x 213,083 GBq/a), which are expected to be converted to HTO in ground-level air.

predictions were within a factor of 2 of the measured concentrations. Performance might be improved by representing plume rise, but this is difficult in an annual average model due to seasonal variation in the temperature gradient.

These results are consistent with observations by Chouhan and Davis (2001) that steady-state Gaussian model predictions are within a factor of 2 of measured annual average concentrations at a majority of monitoring stations around nuclear power plants, at distances of 700 m to 3 km. These power plant data suggested over-prediction by approximately 50% on average.

Tritium concentrations in soil water were modelled from annual emissions to air for 2005 (270,000 GBq/a) and 2006 (74,000 GBq/a). The modelled air concentrations for 2006 were discussed above. The partitioning from air to soil water was modelled as described in Section 4.1.1 (Equation 4.1). Tritium in soil water (sample depth 10 cm) was measured by AECL in September 2006 and September 2007. The one year off-set in soil monitoring data reflects anticipated lag in equilibration of the soil compartment. Emissions declined sharply over the 2005-2007 period, and were only 6,348 GBq/a in 2007.

Model predictions for soil water were made at monitoring well locations, and these were matched with nearby soil sampling locations. All the soil sampling locations were within 100 m of the stack (Figure 5.3). Table 5.2 shows the modelled and measured concentrations of tritium in the soil water. The concentrations predicted from 2006 emissions were in the same range as concentrations measured in 2007. The majority (82%) of model predictions were within a factor of 2 of measured concentrations. The predicted/measured ratio ranged from 0.31 to 2.32 and averaged 1.35. Thus on average, predictions were approximately 35% greater than measured values, assuming a soil lag time of approximately one year.

It is important to note that the soil model in Section 4.1.1 is not time-dependent. It incorporates no lag time. It assumes soil in equilibrium with air on an average annual basis. During periods of strongly decreasing concentration in air, this model will tend to under-predict the soil water (compare 2006 modelled to 2006 measured in Table 5.2). Conversely, during periods of strongly increasing concentration in air the model will tend to over-predict the soil water.

The degree of lag time observed in the soil water compartment will depend on the depth of soil sampled and on site-specific factors that determine the net infiltration rate and hence the residence time of soil water in the shallow soil zone. These factors may vary substantially among sampling stations as a function of soil texture, compaction, and vegetation characteristics.

Soil water samples are substantially elevated in the immediate vicinity of the stack (SS14, SS15, SS16, SS29, SS30, SS31). Roof run-off is also released to this area. Tritium in roof run-off averaged 5,107 Bq/L in 2006. Run-off concentrations during processing averaged 15,288 (Table 5.3). Based on the greater emissions in 2005 roof run-off may have been on the order of 18,600 Bq/L on an annual average basis. Higher concentrations in soil water in

the immediate vicinity of the stack are likely attributable to washout and/or condensation running down the stack wall. Samples of these stack drippings during precipitation events in 2006 averaged 2,298,133 Bq/L during processing and 3,000 Bq/L at other times.

Tritium concentrations in groundwater at monitoring well locations were predicted based on the history of facility emissions from 1991 to 2006. Model predictions for wells were compared to October 2007 measurements of tritium in groundwater.

The first step in predicting well water concentrations is to model the air and soil water concentrations at each well location for each year of emission. Air and soil water concentrations were predicted using the IMPACT model. The emissions and modelled soil water concentrations for each well in each year are listed in Table 5.4.

The second step in predicting well water concentrations is to estimate groundwater travel time from the shallow soil to the well screen depth, and allow for decay of the tritium in soil water while it travels vertically down to the well screen. This is the model described in Section 4.1.1 (Equation 4.2). Using a vertical infiltration rate of 0.5 m/a, the date of soil water origin was determined for each well, as shown in Table 5.5. The Modelled soil water concentration for that date (from Table 5.4) is also shown, along with the predicted concentration after decay during transit to the well screen. This predicted concentration in well water is compared to the 2007 measured concentration.

The predicted/measured ratio ranged from 0.07 to 95, with a geometric mean of 1.52. Approximately 65% of wells were predicted within a factor of 3 of the measured values. Of the 8 predictions that fell outside this range, 7 were over-predictions and 1 was an under-prediction. The under-prediction occurred at MW07-18 which is close to the stack and was likely influenced by snow melt due to snow storage in this area.

Of the 7 over-predicted well water concentrations, 6 of the wells were screened at the bedrock surface, and were likely subject to the effects of horizontal groundwater flow along the bedrock surface. This was suggested by tritium analysis of the soil water in the drill cuttings, which had higher tritium concentrations than those subsequently measured in well water. The soil core profile for MW07-21 (Figure 5.4) illustrates this pattern.

Figure 5.5 is a plot of modelled vs. measured concentrations of tritium in well water. Overall, there is reasonable agreement, with larger discrepancies at a few wells. The discrepancies are generally explainable in terms of sources or aquifer features not represented in the model.

The groundwater vertical velocity likely varies among locations based on subsurface soil conditions. While 0.5 m/a provides a good fit to well data at most locations, higher velocities are suggested for some wells screened in bedrock. For example, tritium concentrations in MW07-28, MW07-29 and MW07-30 could not have originated in the early 1980's prior to facility operation, as calculated from the 0.5 m/a velocities. A velocity closer to 0.9 m/a is indicated for these wells.

Table 5.1: Modelled and Measured Tritium in Air at SRBT

Location	Tritium in Air (Bq/m³)		Model /Measured
	Measured	Modelled	
1	11.42	19.33	1.69
2	6.31	12.01	1.90
3	2.48	4.54	1.83
4	19.05	46.31	2.43
5	7.07	10.80	1.53
6	3.42	7.86	2.30
7	2.20	3.14	1.42
8	15.96	8.16	0.51
9	9.11	7.50	0.82
10	5.85	3.19	0.54
11	7.56	23.43	3.10
12	1.16	3.29	2.83
13	0.57	1.11	1.94
14	0.39	0.97	2.49
15	15.12	21.45	1.42
16	3.47	5.27	1.52
17	0.28	0.68	2.42
18	30.65	20.10	0.66
19	13.81	14.14	1.02
20	4.10	5.70	1.39
21	1.77	1.80	1.02
22	21.45	26.97	1.26
23	5.99	17.70	2.95
24	3.33	9.91	2.98
25	26.14	42.32	1.62
26	6.48	21.75	3.36
27	3.43	6.24	1.82
28	1.62	2.16	1.33
29	24.21	65.66	2.71
30	33.24	96.14	2.89
31	35.66	41.49	1.16

See Figure 5.1 for locations of monitoring stations

Table 5.2: Modelled and Measured Tritium in Soil Water at SRBT

Sampling Location	Measured (Bq/L)			Modelled (Bq/L)[1]		Modelled Location	06Model / 07Measured
	2006	2007[2]		2005	2006		
SRB-SS1	2,374	1,218	L	-	-		
SRB-SS2	1,634	1,340	L	5,738	1015	MW06-1	0.76
SRB-SS3	1,004	535	L	3,108	369	MW07-12	0.69
SRB-SS4	804	566	L	5,271	902	MW07-31	1.59
SRB-SS5	1,598	599	L	5,738	1015	MW06-1	1.69
SRB-SS6	1,293	592	L	3,108	369	MW07-12	0.62
SRB-SS7	1,285	706	L	5,271	902	MW07-31	1.28
SRB-SS8	1,580	1,752	L	-	-		
SRB-SS9	2,080	1,694		-	-		
SRB-SS10	19,734	3,807		-	-		
SRB-SS10-A	8,923	913	L	-	-		
SRB-SS11	20,113	901	L	-	-		
SRB-SS12	15,344	6,036		-	-		
SRB-SS13	13,277	15,202		-	-		
SRB-SS14	31,353	60,450		-	-		
SRB-SS15	248,602	39,877		-	-		
SRB-SS16	366,747	17,115		-	-		
SRB-SS17	3,831	1,053		-	-		
SRB-SS18	1,439	961		1,719	293	MW07-26	0.31
SRB-SS19	695	705		-	-		
SRB-SS20	1854	746		-	-		
SRB-SS21	684	834		-	-		
SRB-SS22	1,267	1,026		-	-		
SRB-SS23	-	822	L	-	-		
SRB-SS24	-	612	L	5,309	1,164	MW07-25	1.90
SRB-SS25	-	668	L	-	-		
SRB-SS26	-	610	L	6,533	1,414	MW07-24	2.32
SRB-SS27	-	637		6,147	1,236	MW07-23	1.94
SRB-SS28	-	692		6,147	1,236	MW07-23	1.79
SRB-SS29	-	15,655	-	-			
SRB-SS30	-	1,066,838	-	-			
SRB-SS31	-	16,774	-	-			

[1] *Based on emissions: 2005 - 2.7E14 Bq/a; 2006 - 7.4E13 Bq/a*
[2] *L= less than*

Table 5.1: Modelled and Measured Tritium in Air at SRBT

Location	Tritium in Air (Bq/m³)		Model /Measured
	Measured	Modelled	
1	11.42	19.33	1.69
2	6.31	12.01	1.90
3	2.48	4.54	1.83
4	19.05	46.31	2.43
5	7.07	10.80	1.53
6	3.42	7.86	2.30
7	2.20	3.14	1.42
8	15.96	8.16	0.51
9	9.11	7.50	0.82
10	5.85	3.19	0.54
11	7.56	23.43	3.10
12	1.16	3.29	2.83
13	0.57	1.11	1.94
14	0.39	0.97	2.49
15	15.12	21.45	1.42
16	3.47	5.27	1.52
17	0.28	0.68	2.42
18	30.65	20.10	0.66
19	13.81	14.14	1.02
20	4.10	5.70	1.39
21	1.77	1.80	1.02
22	21.45	26.97	1.26
23	5.99	17.70	2.95
24	3.33	9.91	2.98
25	26.14	42.32	1.62
26	6.48	21.75	3.36
27	3.43	6.24	1.82
28	1.62	2.16	1.33
29	24.21	65.66	2.71
30	33.24	96.14	2.89
31	35.66	41.49	1.16

See Figure 5.1 for locations of monitoring stations

Table 5.2: Modelled and Measured Tritium in Soil Water at SRBT

Sampling Location	Measured (Bq/L)			Modelled (Bq/L)[1]		Modelled Location	06Model / 07Measured
	2006	2007[2]		2005	2006		
SRB-SS1	2,374	1,218	L	-	-		
SRB-SS2	1,634	1,340	L	5,738	1015	MW06-1	0.76
SRB-SS3	1,004	535	L	3,108	369	MW07-12	0.69
SRB-SS4	804	566	L	5,271	902	MW07-31	1.59
SRB-SS5	1,598	599	L	5,738	1015	MW06-1	1.69
SRB-SS6	1,293	592	L	3,108	369	MW07-12	0.62
SRB-SS7	1,285	706	L	5,271	902	MW07-31	1.28
SRB-SS8	1,580	1,752	L	-	-		
SRB-SS9	2,080	1,694		-	-		
SRB-SS10	19,734	3,807		-	-		
SRB-SS10-A	8,923	913	L	-	-		
SRB-SS11	20,113	901	L	-	-		
SRB-SS12	15,344	6,036		-	-		
SRB-SS13	13,277	15,202		-	-		
SRB-SS14	31,353	60,450		-	-		
SRB-SS15	248,602	39,877		-	-		
SRB-SS16	366,747	17,115		-	-		
SRB-SS17	3,831	1,053		-	-		
SRB-SS18	1,439	961		1,719	293	MW07-26	0.31
SRB-SS19	695	705		-	-		
SRB-SS20	1854	746		-	-		
SRB-SS21	684	834		-	-		
SRB-SS22	1,267	1,026		-	-		
SRB-SS23	-	822	L	-	-		
SRB-SS24	-	612	L	5,309	1,164	MW07-25	1.90
SRB-SS25	-	668	L	-	-		
SRB-SS26	-	610	L	6,533	1,414	MW07-24	2.32
SRB-SS27	-	637		6,147	1,236	MW07-23	1.94
SRB-SS28	-	692		6,147	1,236	MW07-23	1.79
SRB-SS29	-	15,655	-	-			
SRB-SS30	-	1,066,838	-	-			
SRB-SS31	-	16,774	-	-			

[1] *Based on emissions: 2005 – 2.7E14 Bq/a; 2006 – 7.4E13 Bq/a*
[2] *L= less than*

Table: 5.3: Tritium in Roof Runoff, Stack Drippings and Standing Water near Stacks During Precipitation Events at SRBT

Operating Condition	Roof Runoff [1] (Bq/L)	Stack Drippings [2] (Bq/L)	Standing Water (Bq/L)
Processing	15288	2298133	19339
Not Processing	540	3009	664
2006 Average	5107	-	-

[1] *April - August, 2006*

[2] *October - December, 2006*

Table 5.4: Modelled Tritium in Soil Water at Well Locations at SRBT in Previous Years

Year	Emissions (G-Bq/a)	Roof Runoff (Bq/L)	MW86-1 MW07-13 MW07-38	MW86-2 MW07-11	MW86-3	MW86-8 MW07-12	MW86-9 MW07-15	MW86-10	MW07-14	MW07-16 MW07-17	MW07-18 MW07-29 MW07-34	MW07-19	MW07-20	MW07-21	MW07-22 MW07-31	MW07-23	MW07-24 MW07-32	MW07-25 MW07-33	MW07-26	MW07-27 MW07-28 MW07-35	MW07-36	MW07-37
1991	160,000	23,250	4,557	1,225	2,151	3,016	273	85,076	2,614	2,120	5,636	4,030	1,925	3,296	4,425	4,698	4,736	3,779	1,737	5,195	6,197	3 705
1992	260,000	37,968	7,441	2,001	3,512	4,924	446	138,931	4,269	3,462	9,203	6,582	3,143	5,382	7,226	7,672	7,734	6,171	2,836	8,484	10,119	6,050
1993	150,000	22,311	4,373	1,176	2,064	2,894	262	81,640	2,509	2,034	5,408	3,868	1,847	3,163	4,246	4,508	4,545	3,627	1,666	4,986	5,946	3,555
1994	220,000	32,823	6,433	1,730	3,036	4,257	385	120,104	3,691	2,992	7,956	5,690	2,717	4,653	6,247	6,633	6,686	5,335	2,452	7,335	8,748	5,230
1995	260,000	38,189	7,484	2,013	3,533	4,953	448	139,740	4,294	3,482	9,257	6,620	3,162	5,414	7,268	7,717	7,779	6,207	2,852	8,534	10,178	6,085
1996	620,000	90,876	17,810	4,789	8,407	11,787	1,067	332,529	10,218	8,285	22,027	15,753	7,524	12,882	17,296	18,363	18,511	14,771	6,788	20,307	24,220	14,480
1997	1,200,000	134,746	25,798	4,853	11,601	13,972	292	183,077	19,616	15,923	42,462	30,418	10,222	19,042	23,698	27,638	29,370	23,870	7,728	21,849	34,324	18,129
1998	1,600,000	178,212	34,120	6,418	15,343	18,479	386	242,134	25,944	21,060	56,160	40,230	13,520	25,184	31,343	36,554	38,845	31,570	10,221	28,897	45,396	23,976
1999	770,000	86,363	16,535	3,110	7,435	8,955	187	117,339	12,573	10,206	27,215	19,496	6,552	12,204	15,189	17,714	18,824	15,299	4,953	14,004	21,999	11,619
2000	1,700,000	185,512	35,517	6,681	15,971	19,236	402	252,051	27,006	21,922	58,460	41,878	14,074	26,216	32,627	38,051	40,436	32,863	10,640	30,080	47,255	24,958
2001	1,300,000	142,461	27,275	5,130	12,265	14,772	309	193,559	20,739	16,835	44,894	32,160	10,808	20,132	25,055	29,221	31,052	25,237	8,171	23,100	36,289	19,166
2002	930,000	103,913	19,895	3,742	8,946	10,775	225	141,184	15,127	12,280	32,746	23,458	7,883	14,684	18,276	21,314	22,650	18,408	5,960	16,849	26,469	13,980
2003	550,000	61,039	11,686	2,198	5,255	6,329	132	82,932	8,886	7,213	19,235	13,779	4,631	8,626	10,735	12 520	13,304	10,813	3,501	9,897	15,548	8,212
2004	420,000	46,531	8,909	1,676	4,006	4,825	101	63,221	6,774	5,499	14,663	10,504	3,530	6,576	8,184	9,544	10,142	8,243	2,669	7,545	11,853	6,260
2005	270,000	29,970	5,738	1,079	2,580	3,108	65	40,720	4,363	3,542	9,444	6,766	2,274	4,235	5,271	6,147	6,533	5,309	1,719	4,860	7,634	4,032
2006	74,000	5,107	1,015	90	515	369	1	9,338	492	228	700	474	449	943	902	1,236	1,414	1,164	293	643	1,475	592

Table: 5.3: Tritium in Roof Runoff, Stack Drippings and Standing Water near Stacks During Precipitation Events at SRBT

Operating Condition	Roof Runoff [1] (Bq/L)	Stack Drippings [2] (Bq/L)	Standing Water (Bq/L)
Processing	15288	2298133	19339
Not Processing	540	3009	664
2006 Average	5107	-	-

[1] *April - August, 2006*

[2] *October - December, 2006*

Table 5.4: Modelled Tritium in Soil Water at Well Locations at SRBT in Previous Years

Year	Emissions (GBq/a)	Roof Runoff (Bq/L)	MW86-1 MW87-13 MW87-30	MW86-2 MW87-11	MW86-3	MW86-8 MW87-12	MW86-9 MW87-15	MW86-10	MW87-14	MW87-16 MW87-17	MW87-18 MW87-29 MW87-34	MW87-19	MW87-20	MW87-21	MW87-22 MW87-31	MW87-23	MW87-24 MW87-32	MW87-25 MW87-33	MW87-26	MW87-27 MW87-28 MW87-35	MW87-36	MW87-37
1991	160,000	23,250	4,557	1,225	2,151	3,016	273	85,076	2,614	2,120	5,636	4,030	1,925	3,296	4,425	4,698	4,736	3,779	1,737	5,195	6,197	3 705
1992	260,000	37,968	7,441	2,001	3,512	4,924	446	138,931	4,269	3,462	9,203	6,582	3,143	5,382	7,226	7,672	7,734	6,171	2,836	8,484	10,119	6,050
1993	150,000	22,311	4,373	1,176	2,064	2,894	262	81,640	2,509	2,034	5,408	3,868	1,847	3,163	4,246	4,508	4,545	3,627	1,666	4,986	5,946	3,555
1994	320,000	32,823	6,433	1,730	3,036	4,257	385	120,104	3,691	2,992	7,956	5,690	2,717	4,653	6,247	6,633	6,686	5,335	2,452	7,335	8,748	5,230
1995	260,000	38,189	7,484	2,013	3,533	4,953	448	139,740	4,294	3,482	9,257	6,620	3,162	5,414	7,268	7,717	7,779	6,207	2,852	8,534	10,178	6,085
1996	620,000	90,876	17,810	4,789	8,407	11,787	1,067	332,529	10,218	8,285	22,027	15,753	7,524	12,882	17,296	18,363	18,511	14,771	6,788	20,307	24,220	14,480
1997	1,200,000	134,746	25,798	4,853	11,601	13,972	292	183,077	19,616	15,923	42,462	30,418	10,222	19,042	23,698	27,638	29,370	23,870	7,728	21,849	34,324	18,129
1998	1,600,000	178,212	34,120	6,418	15,343	18,479	386	242,134	25,944	21,060	56,160	40,230	13,520	25,184	31,343	36,554	38,845	31,570	10,221	28,897	45,396	23,976
1999	770,000	86,363	16,535	3,110	7,435	8,955	187	117,339	12,573	10,206	27,215	19,496	6,552	12,204	15,189	17,714	18,824	15,299	4,953	14,004	21,999	11,619
2000	1,700,000	185,512	35,517	6,681	15,971	19,236	402	252,051	27,006	21,922	58,460	41,878	14,074	26,216	32,627	38,051	40,436	32,863	10,640	30,080	47,255	24,958
2001	1,300,000	142,461	27,275	5,130	12,265	14,772	309	193,559	20,739	16,835	44,894	32,160	10,808	20,132	25,055	29,221	31,052	25,237	8,171	23,100	36,289	19,166
2002	930,000	103,913	19,895	3,742	8,946	10,775	225	141,184	15,127	12,280	32,746	23,458	7,883	14,684	18,276	21,314	22,650	18,408	5,960	16,849	26,469	13,980
2003	550,000	61,039	11,686	2,198	5,255	6,329	132	82,932	8,886	7,213	19,235	13,779	4,631	8,626	10,735	12,520	13,304	10,813	3,501	9,897	15,548	8,212
2004	420,000	46,531	8,909	1,676	4,006	4,825	101	63,221	6,774	5,499	14,663	10,504	3,530	6,576	8,184	9,544	10,142	8,243	2,669	7,545	11,853	6,260
2005	270,000	29,970	5,738	1,079	2,580	3,108	65	40,720	4,363	3,542	9,444	6,766	2,274	4,235	5,271	6,147	6,533	5,309	1,719	4,860	7,634	4,032
2006	74,000	5,107	1,015	90	515	369	1	9,338	492	228	700	474	419	943	902	1,236	1,414	1,164	293	643	1,475	592

Table 5.5: Modelled and Measured Tritium in Groundwater in SRBT Monitoring Wells

Monitoring Well	Distance from Stacks (m)	Measured Tritium Oct-07 (Bq/L)	Soil Water Origin [1] Modelled Tritium					Model / Measured
			Screen Depth (m)	Years Ago	Date	Soil Water (Bq/L)	Groundwater after Decay (Bq/L)	
MW06-1 Clay	50	43,586	4.34	8.68	1998	34120	20903	0.48
MW06-2 Clay	75	4,050	4.54	9.08	1998	6418	3844	0.95
MW06-3 Clay	50	3,139	5.36	10.72	1996	8407	4590	1.46
MW06-8 Clay	55	311	5.98	11.96	1995	4953	2522	8.11
MW06-9 Clay	25	1,489	4.64	9.28	1998	386	229	0.15
MW06-10 BRS	0	29,795	6.69	13.38	1994	120104	56434	1.89
MW07-11 BRS	75	400	6.31	12.62	1994	1730	849	2.12
MW07-12 BRS	55	194	6.61	13.22	1994	4257	2018	10.40
MW07-13 BRS	50	10,057	5.72	11.44	1996	17810	9337	0.93
MW07-14 BRS	40	2,357	6.4	12.80	1994	3691	1792	0.76
MW07-15 BRS	25	227	6.32	12.64	1994	385	189	0.83
MW07-16 BRS	15	6,646	6.23	12.46	1995	3482	1723	0.26
MW07-17 BR	15	103	13.7	27.40	1980	-	-	-
MW07-18 BRS	10	58,139	6.38	12.76	1994	7956	3871	0.07
MW07-19 BRS	20	4,229	6.59	13.18	1994	5690	2704	0.64
MW07-20 BRS	90	628	6.97	13.94	1993	1847	841	1.34
MW07-21 BRS	110	102	6.85	13.70	1993	3163	1460	14.31
MW07-22 BRS	70	557	6.27	12.54	1994	6247	3078	5.53
MW07-23 BRS	90	595	5.16	10.32	1997	29370	16402	27.57
MW07-24 BRS	115	102	5.72	11.44	1996	18511	9704	95.14
MW07-25 BRS	105	1,230	5.93	11.86	1995	6207	3178	2.58
MW07-26 BRS	50	2,731	6.74	13.48	1994	2452	1146	0.42
MW07-27 BRS	55	6,959	7.46	14.92	1992	8484	3654	0.53
MW07-28 BR	55	8,088	13.73	27.46	1980	-	-	-
MW07-29 BR	10	38,797	12.48	24.96	1982	-	-	-
MW07-30 BR	50	14,185	13.22	26.44	1981	-	-	-
MW07-31 BR	70	801	12.46	24.92	1982	-	-	-
MW07-32 BR	115	143	12.46	24.92	1982	-	-	-
MW07-33 BR	105	678	13.61	27.22	1980	-	-	-
MW07-34 BR	10	45,544	8.7	17.40	1990	-	-	-
MW07-35 BR	55	14,824	8.66	17.32	1990	-	-	-
MW07-36 BR	80	9,100	8.54	17.08	1990	-	-	-
MW07-37 BR	60	3,297	8	16.00	1991	3705	1502	0.46

BR= Bedrock, BRS= Bedrock Surface

[1] date of origin assumes a vertical velocity of 0.5 m a; this is a typical value but may vary among locations based on subsurface soil conditions.

Table 5.5: Modelled and Measured Tritium in Groundwater in SRBT Monitoring Wells

Monitoring Well	Distance from Stacks (m)	Measured Tritium Oct-07 (Bq/L)	Soil Water Origin[1] Modelled Tritium					
			Screen Depth (m)	Years Ago	Date	Soil Water (Bq/L)	Groundwater after Decay (Bq/L)	Model / Measured
MW06-1 Clay	50	43,586	4.34	8.68	1998	34120	20903	0.48
MW06-2 Clay	75	4,050	4.54	9.08	1998	6418	3844	0.95
MW06-3 Clay	50	3,139	5.36	10.72	1996	8407	4590	1.46
MW06-8 Clay	55	311	5.98	11.96	1995	4953	2522	8.11
MW06-9 Clay	25	1,489	4.64	9.28	1998	386	229	0.15
MW06-10 BRS	0	29,795	6.69	13.38	1994	120104	56434	1.89
MW07-11 BRS	75	400	6.31	12.62	1994	1730	849	2.12
MW07-12 BRS	55	194	6.61	13.22	1994	4257	2018	10.40
MW07-13 BRS	50	10,057	5.72	11.44	1996	17810	9337	0.93
MW07-14 BRS	40	2,357	6.4	12.80	1994	3691	1792	0.76
MW07-15 BRS	25	227	6.32	12.64	1994	385	189	0.83
MW07-16 BRS	15	6,646	6.23	12.46	1995	3482	1723	0.26
MW07-17 BR	15	103	13.7	27.40	1980	-	-	-
MW07-18 BRS	10	58,139	6.38	12.76	1994	7956	3871	0.07
MW07-19 BRS	20	4,229	6.59	13.18	1994	5690	2704	0.64
MW07-20 BRS	90	628	6.97	13.94	1993	1847	841	1.34
MW07-21 BRS	110	102	6.85	13.70	1993	3163	1460	14.31
MW07-22 BRS	70	557	6.27	12.54	1994	6247	3078	5.53
MW07-23 BRS	90	595	5.16	10.32	1997	29370	16402	27.57
MW07-24 BRS	115	102	5.72	11.44	1996	18511	9704	95.14
MW07-25 BRS	105	1,230	5.93	11.86	1995	6207	3178	2.58
MW07-26 BRS	50	2,731	6.74	13.48	1994	2452	1146	0.42
MW07-27 BRS	55	6,959	7.46	14.92	1992	8484	3654	0.53
MW07-28 BR	55	8,088	13.73	27.46	1980	-	-	-
MW07-29 BR	10	38,797	12.48	24.96	1982	-	-	-
MW07-30 BR	50	14,185	13.22	26.44	1981	-	-	-
MW07-31 BR	70	801	12.46	24.92	1982	-	-	-
MW07-32 BR	115	143	12.46	24.92	1982	-	-	-
MW07-33 BR	105	678	13.61	27.22	1980	-	-	-
MW07-34 BR	10	45,544	8.7	17.40	1990	-	-	-
MW07-35 BR	55	14,824	8.66	17.32	1990	-	-	-
MW07-36 BR	80	9,100	8.54	17.08	1990	-	-	-
MW07-37 BR	60	3,297	8	16.00	1991	3705	1502	0.46

BR= Bedrock, BRS= Bedrock Surface

[1] date of origin assumes a vertical velocity of 0.5 m/a; this is a typical value but may vary among locations based on subsurface soil conditions.

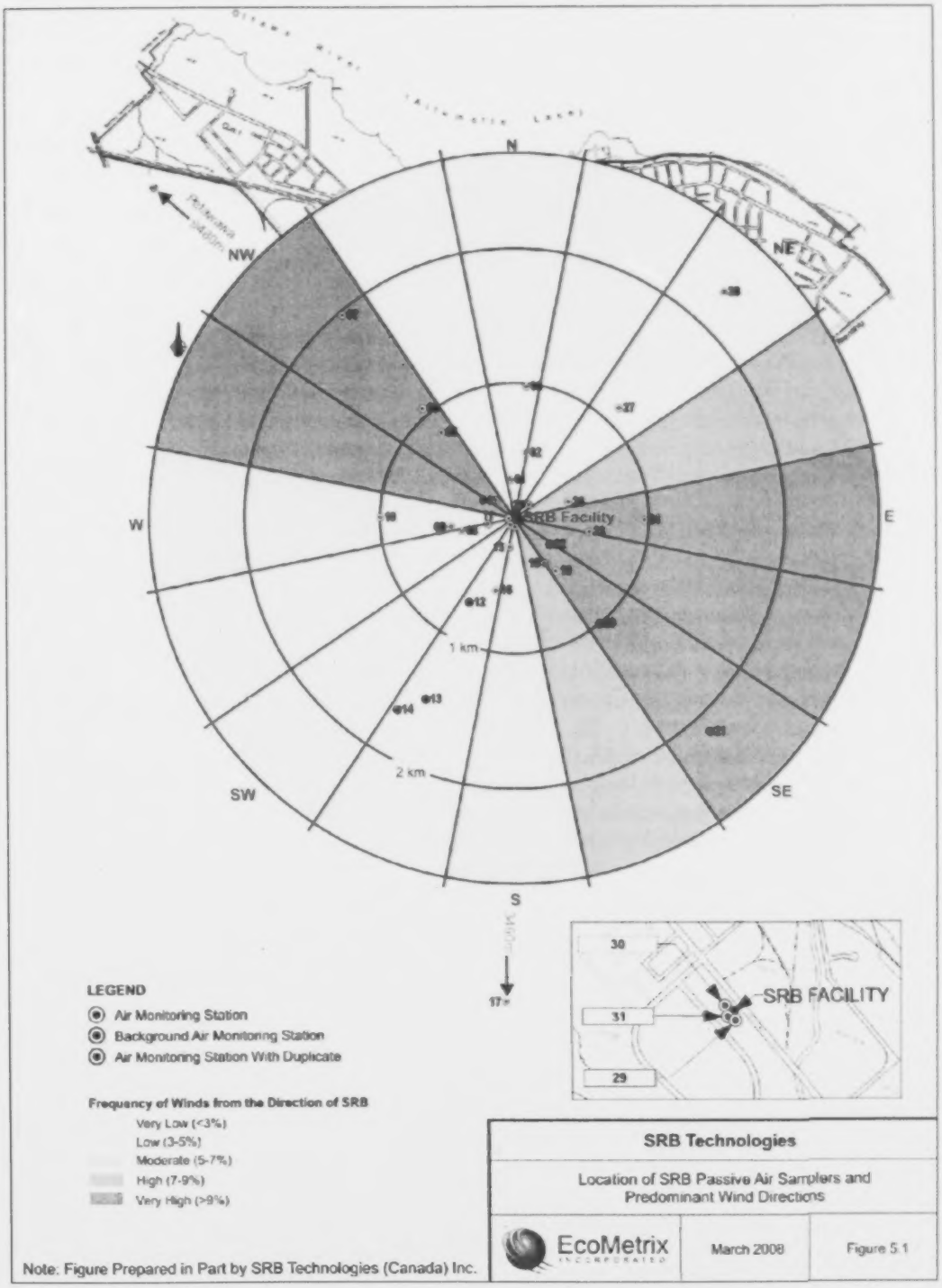

N
NE
E
SE
S
SW
W
NW
SRB Facility
1 km
2 km
LEGEND
Air Monitoring Station
Background Air Monitoring Station
Air Monitoring Station With Duplicate
Frequency of Winds from the Direction of SRB
Very Low (<3%)
Low (3-5%)
Moderate (5-7%)
High (7-9%)
Very High (>9%)
30
31
29
SRB FACILITY
SRB Technologies
Location of SRB Passive Air Samplers and Predominant Wind Directions
EcoMetrix
March 2008
Figure 5.1
Note: Figure Prepared in Part by SRB Technologies (Canada) Inc.

Figure 5.2: Modelled vs Measured Tritium in Air at SRBT, 2006

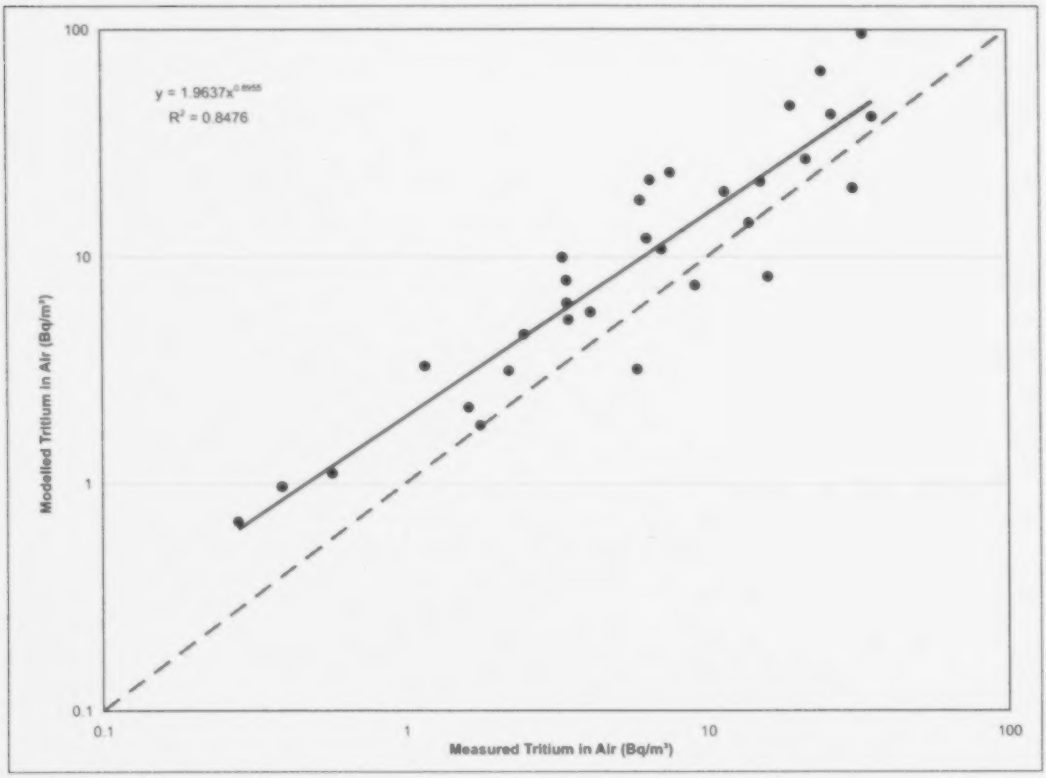


Figure 5.3: Soil Water and Groundwater Sampling Locations at SRBT

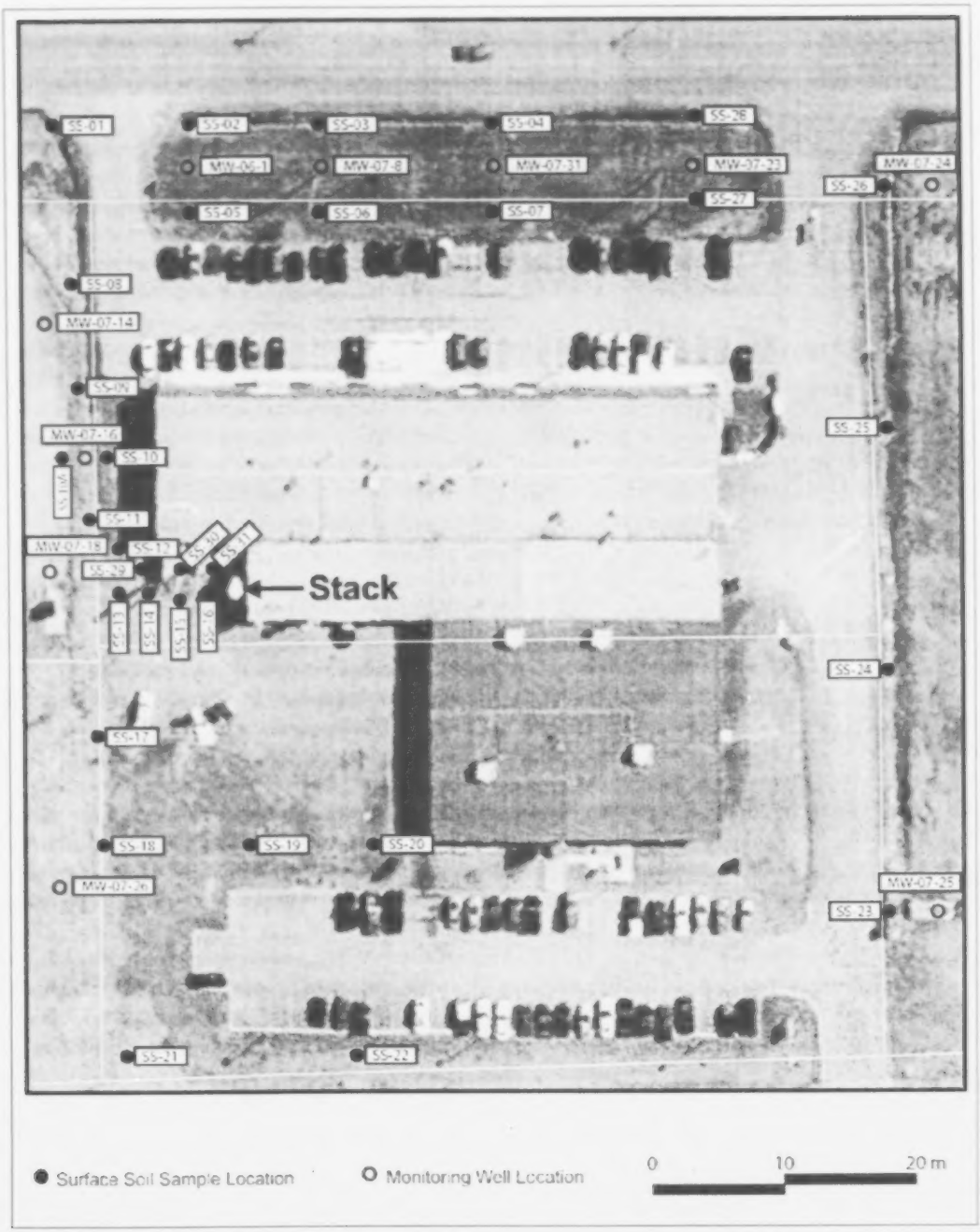


Figure 5.3: Soil Water and Groundwater Sampling Locations at SRBT

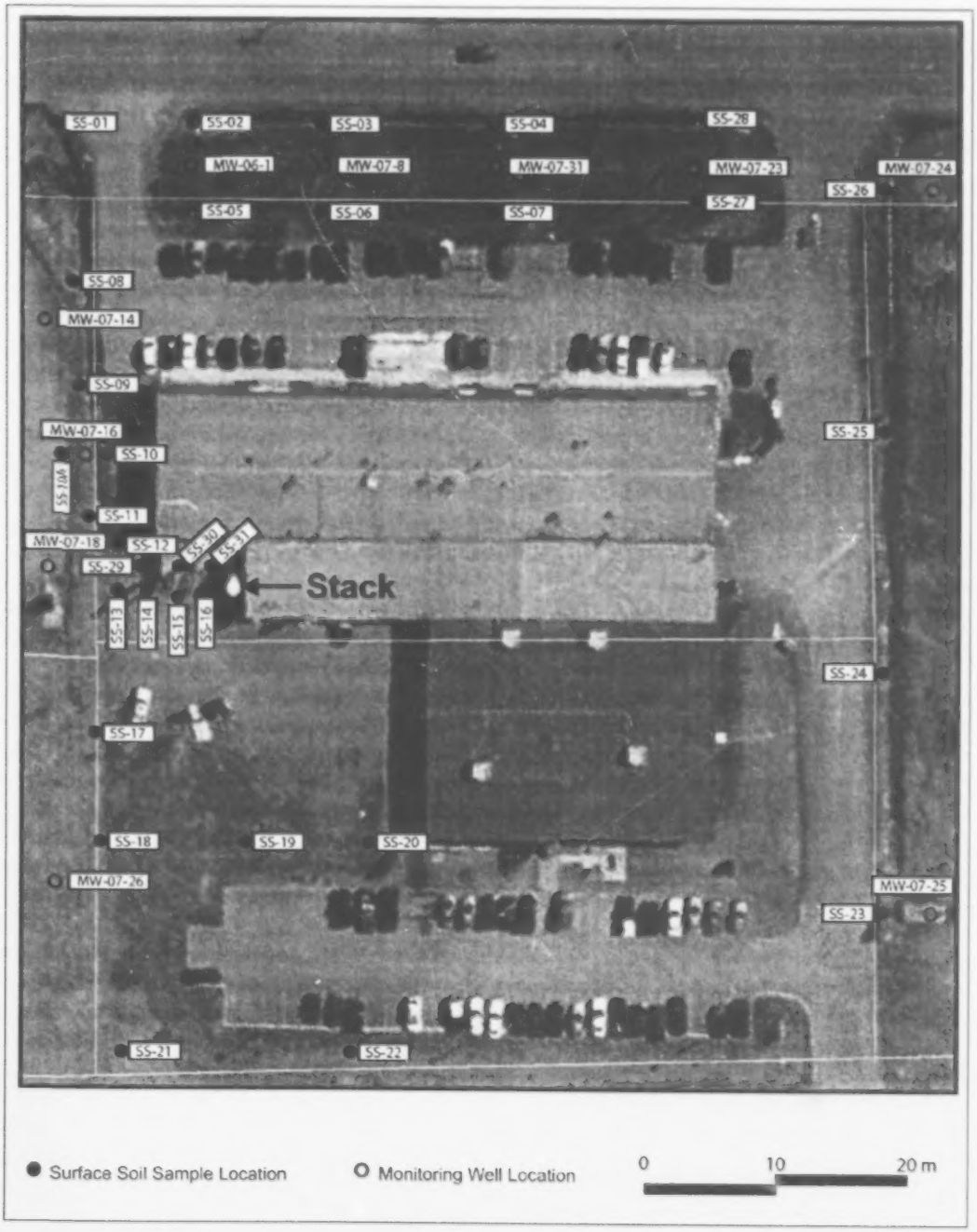


Figure 5.4: Tritium Concentrations in Soil Water and Groundwater at MW07-21 on the SRBT Site

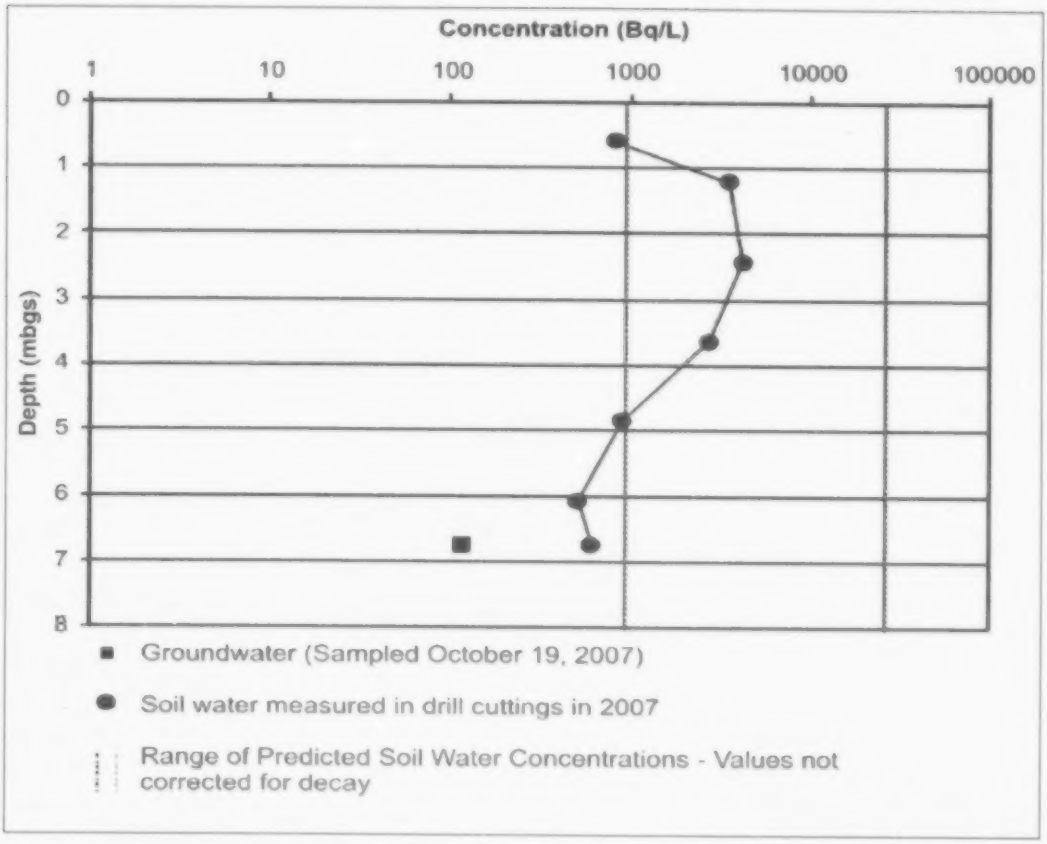
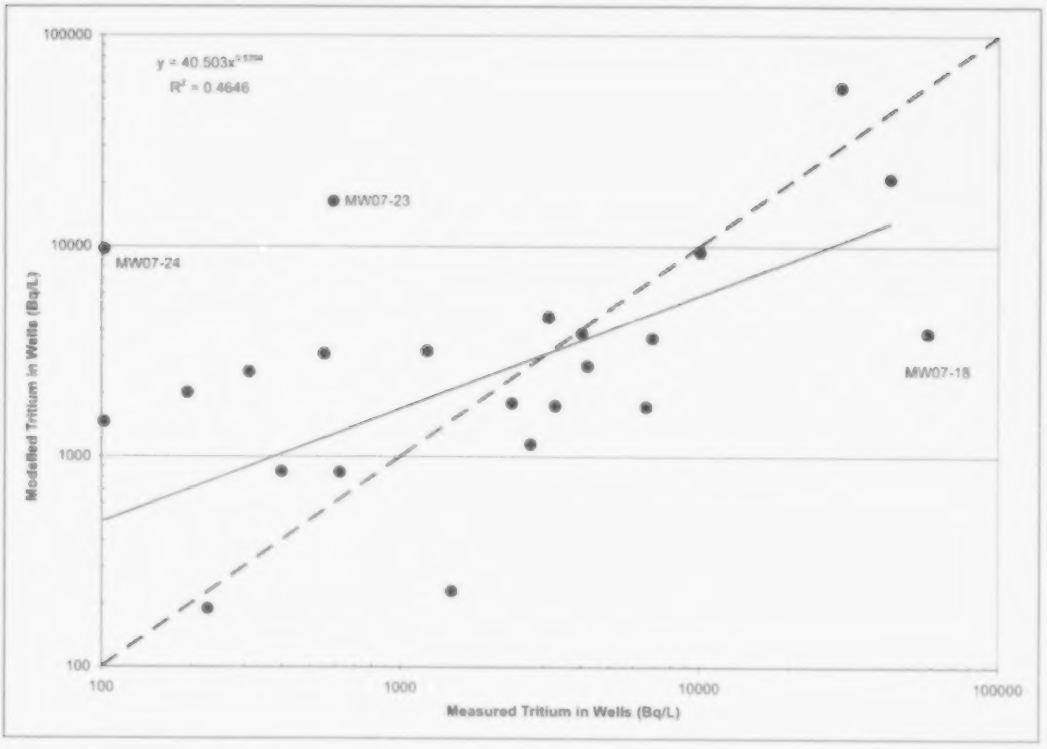


Figure 5.5: Modelled vs Measured Tritium in SRBT Wells, 2007



Outliers at MW07-23, MW07-24 and MW07-18 omitted from the fit line

5.1.2 Shield Source Incorporated

Modelling of the SSI facility was undertaken based on information in its 2007 Annual Compliance Report (SSI, 2008). The report presents facility emissions as well as air and surface water monitoring data for locations around the facility, at distances from 74 m to several kilometres.

The present study is focused on tritium (HTO) in air and pond or marsh water around the facility. Rivers and creeks are also monitored, but are not expected to equilibrate with air during their brief period of transit across the area of exposure to facility emissions. Some private wells are also monitored; however, well depths are not reported. Depth information is required for modelling of wells.

Meteorological data for Peterborough for 2007 were obtained from Trent University. These data contain the standard deviation in wind direction, which is required for calculation of stability class and production of a triple joint frequency (TJF) distribution. The TJF was determined and used to model tritium in air around the facility. The stack height and diameter were 9.1 m and 0.5 m, respectively. The stack exit velocity was 8.2 m/s, based on measured air flow of 3,415 cfm. The gas temperature was assumed to be only 1° C above ambient (i.e., no thermal plume rise). A 7 m height was assumed for adjacent buildings.

Tritium in air concentrations were modelled using the IMPACT code, which contains a steady-state sector-averaged Gaussian plume model, as described in the CSA (2008) N288.1 guidelines. The model used 2007 annual emissions (24,900 GBq/a)[2] and 2007 meteorological data. As a test of the air model, the predicted ground level concentrations of tritium for 2007 were compared to annual average measured concentrations for the same year at 16 locations around the site. The air monitoring was conducted using passive samplers, which were analyzed by Monserco Limited.

The modelled vs. measured air concentrations are tabulated in Table 5.6 and illustrated in Figure 5.6. The results indicate that the air model generally over-predicts tritium in near-field air (usually by 2 to 4 fold), but generally agrees with measurements (within a factor of 2) at distances beyond 1 km (Figure 5.7). Near-field over prediction in this example may result from not considering thermal plume rise. This is a seasonally variable phenomenon.

The modelled vs. measured pond water concentrations are tabulated in Table 5.7 and illustrated in Figure 5.8. The results indicate reasonable model agreement with measured pond water. The majority (58%) of predicted concentrations were within a factor of 2 of measured concentrations, and almost all (92%) were within a factor of 3. The predicted/measured ratio ranged from 0.37 to 3.66 and averaged 1.28. The near-field ponds (W3, WG5, W4) are of particular interest because, while tritium in air was

[2] Includes HTO emissions, plus 2% of HT emissions (0.02 x 84,700 GBq/a) which are expected to be converted to HTO in ground-level air.

over-predicted at these locations (by factors of 3 to 4), the modelled pond water was somewhat under-predicted (within a factor of 2 or 3). Possibly, near-field ponds show the influence of higher air concentrations in previous years, as reflected in up-gradient hydrological inputs, whereas the model assumes instant equilibrium between air and pond water.

Table 5.6: Modelled and Measured Tritium in Air at SSI in 2007

Location	Distance (m)	Measured (Bq/m^3)	Modelled (Bq/m^3)	Model / Measured
A1	74	7.22	22.72	3.15
A2	240	1.43	6.18	4.32
A3	210	3.05	7.35	2.41
A4	250	3.37	8.99	2.67
A5	220	2.96	10.24	3.46
A6	210	1.17	12.33	10.5
A7	200	1.2	4.02	3.35
A8	870	0.59	1.47	2.49
A9	1,500	0.49	0.93	1.89
A10	1,500	1.08	0.94	0.87
A11	1,200	1	1.41	1.41
A12	1,000	0.65	0.61	0.94
A13	1,000	0.53	0.82	1.54
A14	1,000	0.48	1.74	3.63
A15	2,500	0.39	0.36	0.92

Table 5.7: Modelled and Measured Tritium in Ponds at SSI in 2007

Location	Distance (m)	Measured (Bq/m3)	Modelled (Bq/m3)	Model / Measured
W3	170	1,872	1,101	0.59
W4	250	1,088	870.6	0.80
W6	210	652	1,162	1.78
W8	870	148	145.0	0.98
W9	1,500	82	92.58	1.13
W10	1,500	227	92.60	0.41
W11	1,200	50	140.5	2.81
W12	1,000	125	60.93	0.49
W13	1,200	50	80.82	1.62
W14	1,000	47	172.1	3.66
W15	2,500	50	36.54	0.73
WG5	220	1,778	658.8	0.37

Figure 5.6: Modelled vs Measured Tritium in Air at SSI – 2007 Data

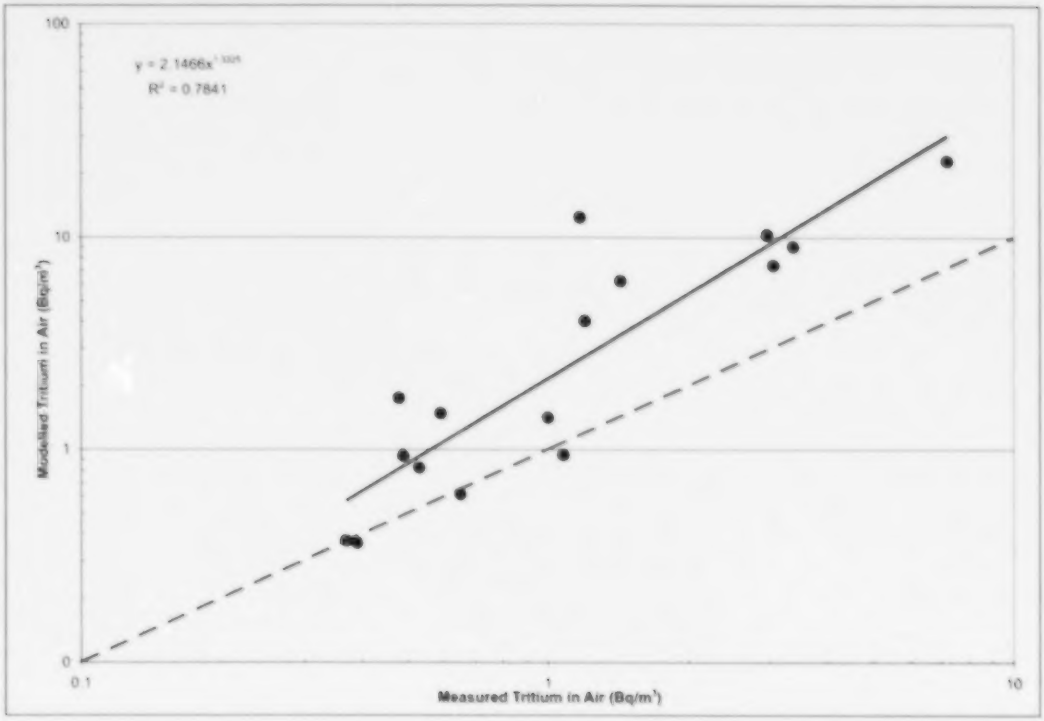


Figure 5.7: Ratio of Modelled to Measured Air vs Distance at SSI – 2007 Data

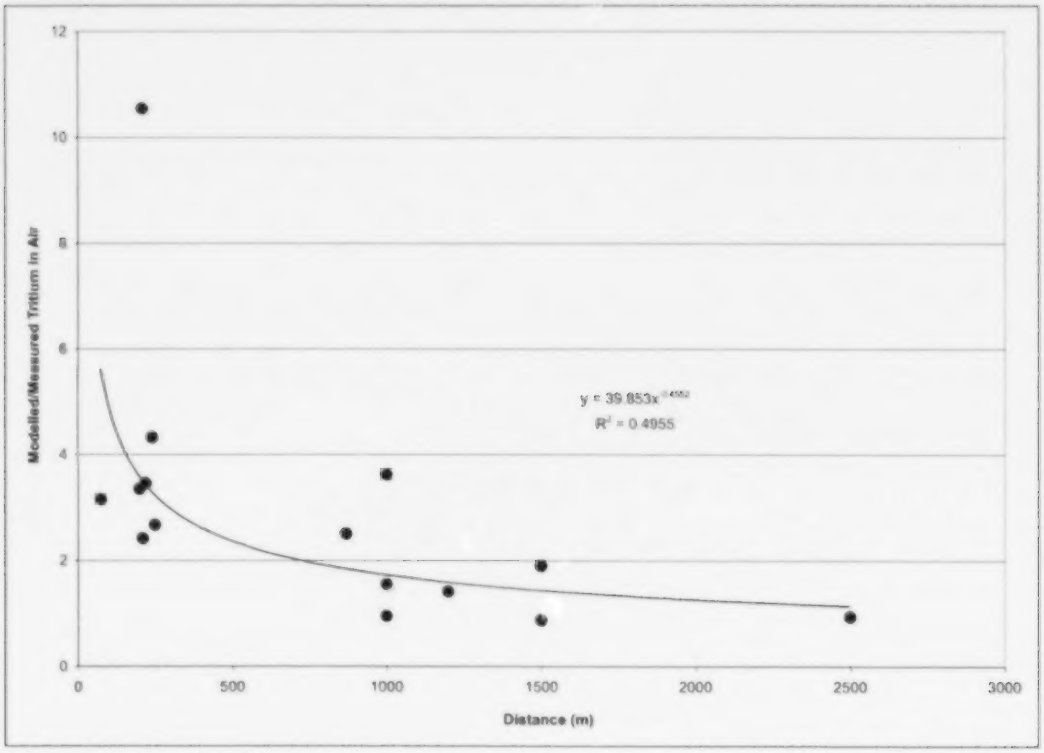
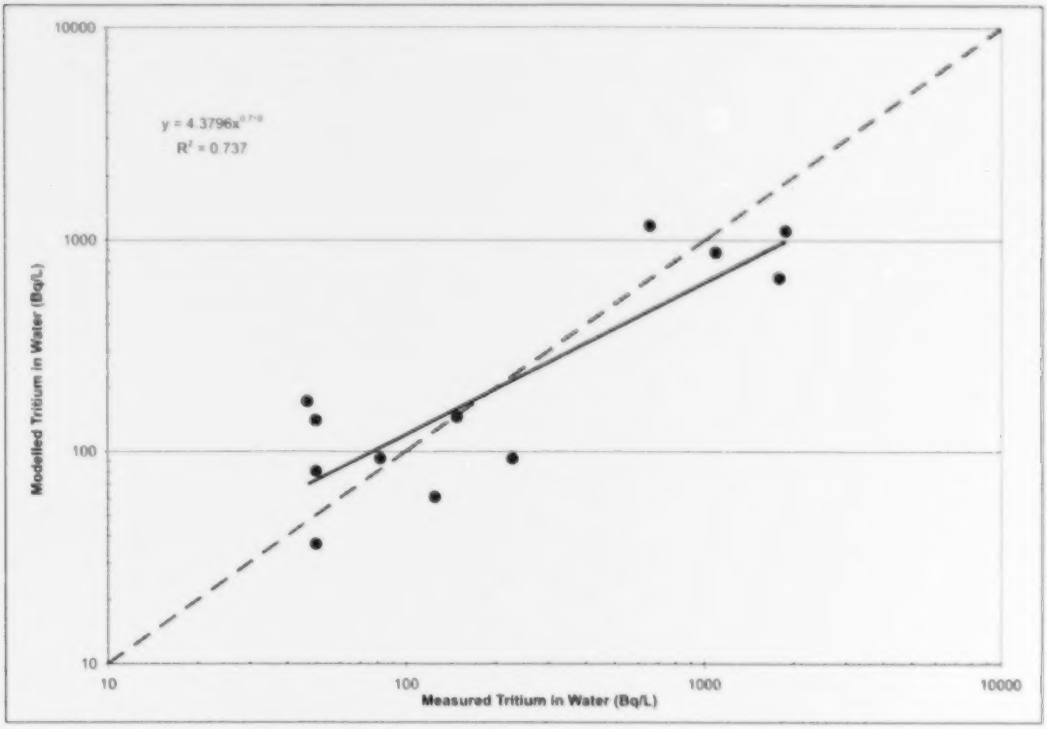


Figure 5.8: Modelled vs Measured Tritium in Ponds at SSI in 2007



5.2 Nuclear Power Stations

Three nuclear generating stations are considered in this section. The Pickering Nuclear (PN) and Darlington Nuclear (DN) facilities are operated by Ontario Power Generation (OPG) on the north shores of Lake Ontario. The Bruce Nuclear (BN) facility is operated by Bruce Power near Tiverton, Ontario, on the north shore of Lake Huron. Readily available data pertinent to annual average dispersion and environmental partitioning of tritium were utilized. An annual timeframe is appropriate given interest in annual doses.

5.2.1 Pickering (PN)

Modelling of the PN facility was undertaken based on information in OPG's 2007 Results of Radiological Environmental Monitoring Programs (Borromeo, 2008). The report presents facility emissions as well as air and precipitation monitoring data for locations around the facility. Air and precipitation monitoring stations include various perimeter sites within 2 km of the facility, as well as more distant sites as far away as 10.4 km from the facility.

Both active and passive air samplers are used. Samples are collected quarterly and averaged to obtain an annual average value. Passive samplers at OPG generally give results that are twice the active sampler results[3]. Since only passive samplers are located in the far-field, estimates were made of passive sampler results at some perimeter locations where only active samplers exist.

Tritium concentrations in air were modelled using the IMPACT code, which contains a steady-state sector-averaged Gaussian plume model, as described in the CSA (2008) N288.1 guidelines. The model used 2007 annual emissions (560 TBq/a HTO) and 2007 meteorological data. As a test of the air model, the predicted annual average ground level concentrations of tritium for 2007 were compared to annual average measured concentrations for the same year at 6 perimeter locations and several far-field locations.

The modelled vs. measured air concentrations are tabulated in Table 5.8 and illustrated in Figure 5.9. The results indicate that the air model generally predicts tritium within a factor of 2 of measured values (using passive samplers). One out of 9 predictions (11%) fell outside this range. The air concentration was over-predicted at this location (P4). On average, modelled concentrations were approximately 35% higher than measured values.

It should be noted that, if active sampler results are found to be accurate, this would imply that the air model as implemented here is more over-predictive than suggested in Table 5.8, by approximately two-fold.

Soil water is not routinely measured at OPG facilities; however, rain water is a reasonable surrogate measurement, since soil water originates as rain water (with a snowmelt contribution in the spring). The modelled soil water was calculated as described in Section 4.1.1 (Equation 4.1) using an absolute humidity of 0.0089 L/m^3 for the snow-free period (to match the sampled rain events). The modelled and measured values are compared in Table 5.9, and illustrated in Figure 5.10. Six out of 8 predictions (75%) are within a factor of 2 of the measured values, and the predictions are consistently greater than or equal to measured values. On average, modelled concentrations were approximately twice the measured values.

Davis (2006) studied tritium concentrations in air, rain water and soil water at three of the Table 5.8 stations (P2, D8 and F27). He found good agreement between levels of tritium in soil water and those in rain water averaged over the previous five months. The rain/soil water ratio averaged 1.2, ranging from 0.8 to 2. Soil water modelled from air (Equation 4.1) was approximately 50% higher than measured values.

[3] OPG has been using passive sampler results for public dose calculations, as a conservative measure, until it can resolve the differences between the two measurement methods.

Table 5.8: Modelled and Measured Tritium in Air at PN, 2007

Location[1]	HTO Concentration (Bq/m³)			Model / Measured
	Active	Passive	Modelled	
P2	20.8	32.8	28.96	0.88
P3	3.9	7.8[2]	8.73	1.12
P4	1.6	3.2[2]	10.88	3.40
P6	6.8	13.7	16.68	1.22
P10	15.9	31.5	39.12	1.24
P11	3.5	7.0	9.97	1.42
C2	-	6.7	6.21	0.93
DF8	-	1.3	1.00	0.77
F31/F27	-	1.2	1.49	1.24

[1] *1 P2 - P11 are perimeter stations*

[2] *Passive value estimated as 2 x active value*

Table 5.9: Modelled Tritium in Soil Water Compared to Measured Tritium in Rain Water at PN, 2007

Location	HTO Concentration (Bq/L)		
	Rain	Modelled Soil Water	Model/Measured
P2	602	976	1.62
P3	150	294	1.96
P4	91	367	4.03
P6	296	562	1.90
P10	691	1,318	1.91
P11	112	336	3.00
DF8	34	34	1.00
F31/F27	34	50	1.47

Figure 5.9: Modelled vs Measured Tritium in Air at PN, 2007

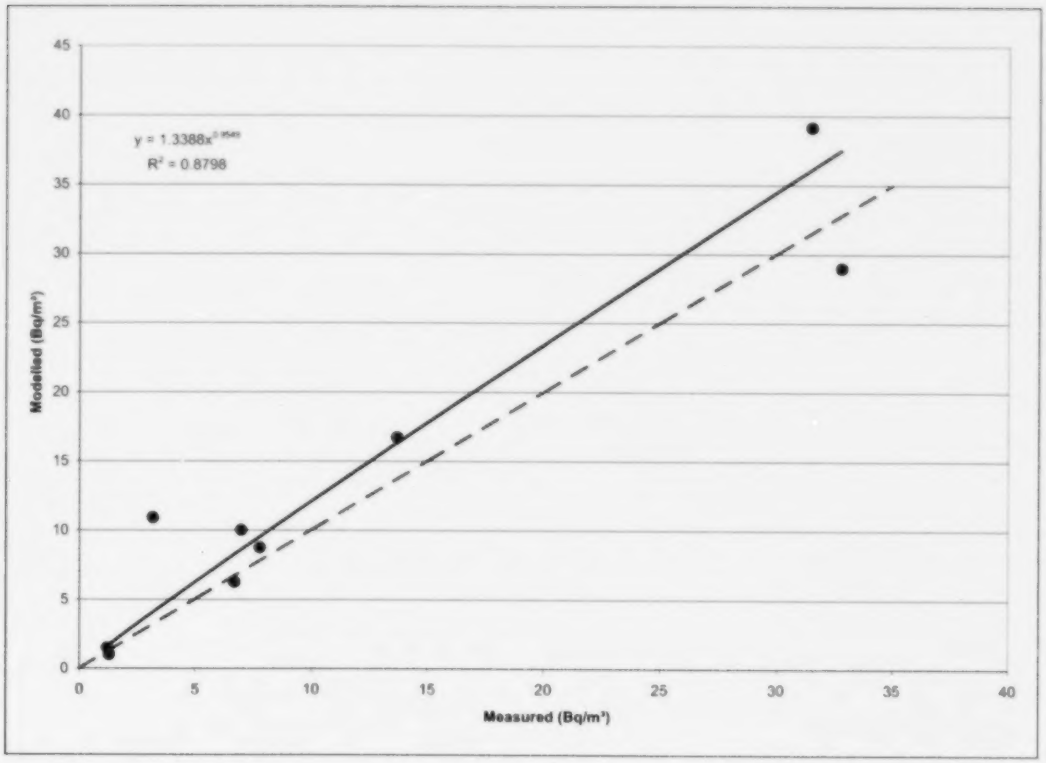
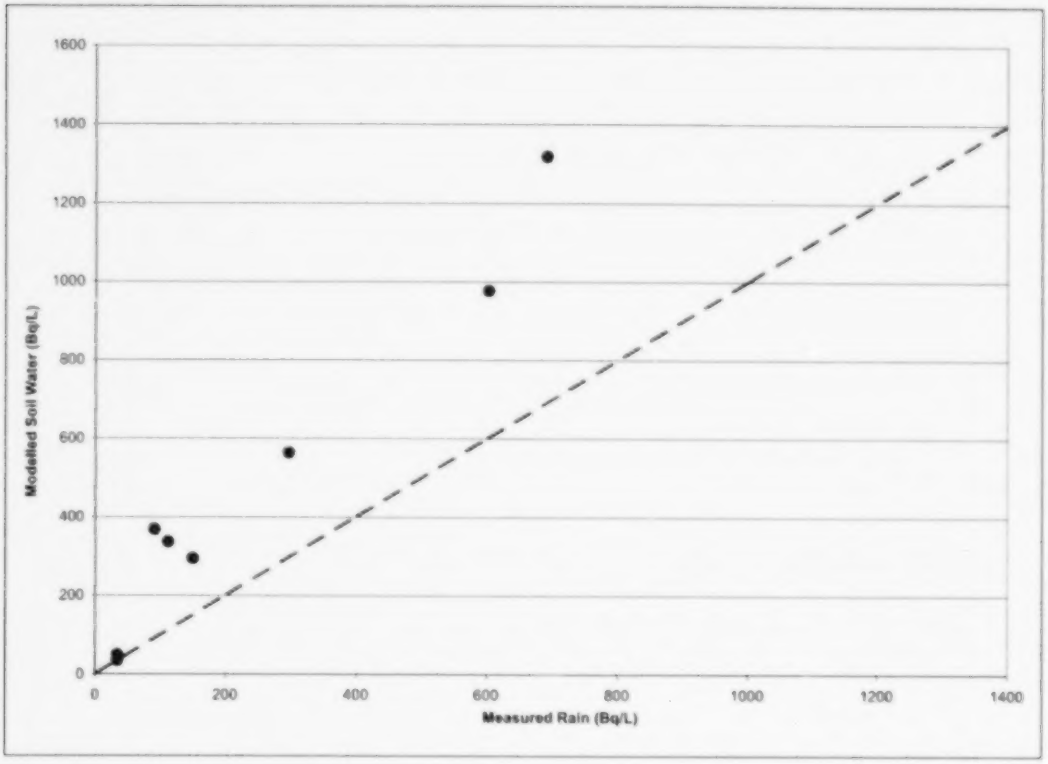


Figure 5.10: Modelled Tritium in Soil Water vs Measured Tritium in Rain Water at PN, 2007



5.2.2 Darlington (DN)

Modelling of the DN facility was undertaken based on information in OPG's 2007 Results of Radiological Environmental Monitoring Programs (Borromeo, 2008). The report presents facility emissions as well as air and precipitation monitoring data for locations around the facility. Air and precipitation monitoring stations include various perimeter sites within 2 km of the facility, as well as more distant sites as far away as 13 km from the facility.

Both active and passive samplers are used. Samples are collected quarterly and averaged to obtain an annual average value. Passive samplers at OPG generally give results that are twice the active sampler results[4] . Since most far-field stations have only passive samplers, estimates were made of passive sampler results at some perimeter locations where only active samplers exist.

[4] OPG has been using passive sampler results for public dose calculations, as a conservative measure, until it can resolve the differences between the two measurement methods.

Tritium (HTO) concentrations in air were modelled using the IMPACT code, which contains a steady-state sector-averaged Gaussian plume model, as described in the CSA (2008) N288.1 guidelines. The model used 2007 annual emissions (160 TBq/a HTO)[4] and 2007 meteorological data. As a test of the air model, the predicted annual average ground level concentrations of tritium for 2007 were compared to annual average measured concentrations for the same year at 6 perimeter locations and 5 far-field locations.

The modelled vs. measured air concentrations are tabulated in Table 5.10 and illustrated in Figure 5.11. The results indicate that model predicted concentrations of HTO in air are generally (91%) within a factor of 2 of measured concentrations (using passive samplers). On average, modelled concentrations were approximately 20% higher than measured values. It should be noted that, if active sampler results are found to be accurate, this would imply that the air model as implemented here is more over-predictive than suggested in Table 5.8, by approximately two-fold.

Table 5.10: Modelled and Measured Tritium in Air at DN, 2007

Location[1]	HTO Concentration (Bq/m³)			Model / Measured
	Active	Passive	Modelled	
D1	1.8	3.7	5.02	1.36
D2	1.6	3.52	3.71	1.06
D3	0.9	1.82	3.04	1.69
D4	0.6	1.2	0.98	0.82
D5	0.5	1.0	0.67	0.67
D6	0.4	0.5	0.33	0.66
D7	0.2	0.4	0.72	1.80
D8	0.6	1.2	0.93	0.78
F1	-	1.1	1.53	1.39
DF5	-	0.2	0.59	2.95
R275	-	1.0	1.34	1.34

[1] *1 P2 - P11 are perimeter stations*

[2] *Passive value estimated as 2 x active value*

[4] Emissions were not adjusted up to account for HTO produced by HT released. 130 TBq/a HT from the tritium removal facility are expected to increase HTO concentrations by approximately 2%.

Table 5.11: Modelled Tritium in Soil Water Compared to Measured Tritium in Rain Water at DN, 2007

Location[1]	HTO Concentration (Bq/L)		Model / Measured
	Rain	Modelled Soil Water	
D1	72	169	2.35
D2	68	125	1.84
D3	27	102	3.78
D4	28	33	1.18
D5	28	23	0.82
D6	20	11	0.55
D7	14	24	1.71
D8	19	31	1.63
F1	42	52	1.24
DF5	13	20	1.54
R275	38	45	1.18

Figure 5.11: Modelled vs Measured Tritium in Air at DN, 2007

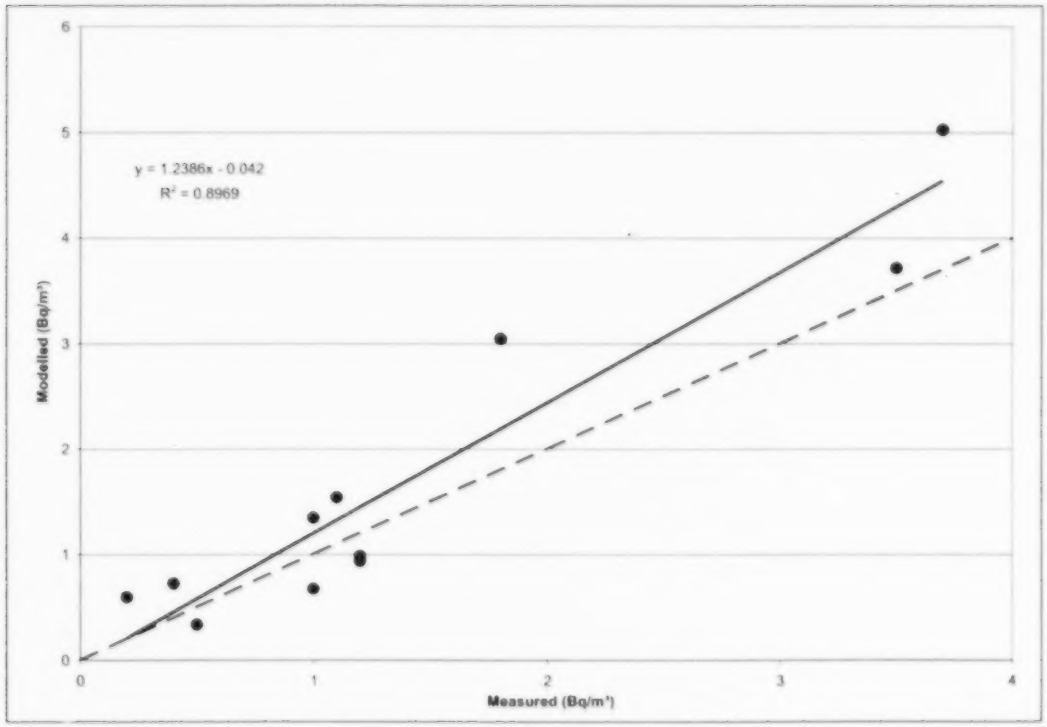
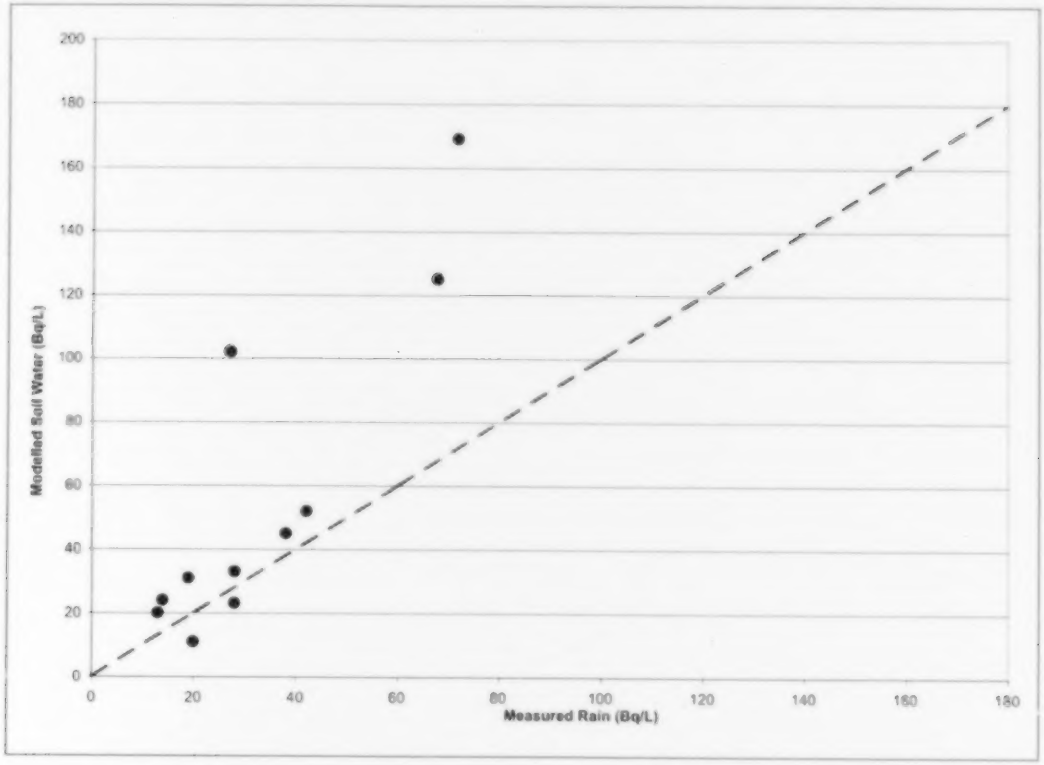


Figure 5.12: Modelled Tritium in Soil Water vs Measured Tritium in Rain Water at DN, 2007



Soil water is not routinely measured at OPG facilities; however, rain water is a reasonable surrogate measurement. The modelled soil water was calculated as described in Section 4.1.1 (Equation 4.1) using an absolute humidity of 0.0089 for the snow-free period (to match the sampled rain events). The modelled and measured values are compared in Table 5.11, and illustrated in Figure 5.12. Nine out of 11 predictions (82%) are within a factor of 2 of the measured values. On average, modelled concentrations were 62% higher than measured values.

5.2.3 Bruce (BN)

Modelling of the BN facility was undertaken based on information in its Annual Summary and Assessment of Environmental Radiological Data for 2006 (McDougall, 2007). The report presents facility emissions as well as air and precipitation monitoring data for locations around the facility. Air monitoring stations include various perimeter sites within 2 km of the facility, as well as more distant sites as far away as 19 km from the facility. Active and passive air samplers are used, and both are present at some perimeter locations. The data for those locations indicate reasonable agreement, and the higher of the two measured values was used. Only passive samplers are utilized in the far-field.

Tritium in air concentrations were modelled using the IMPACT code, which contains a steady-state sector-averaged Gaussian plume model, as described in the CSA (2008) N288.1 guidelines. The model used 2006 annual emissions (951 TBq/a HTO) and 5-year average meteorological data (2002-2006). As a test of the air model, the predicted ground level concentrations of tritium for 2006 were compared to annual average measured concentrations for the same year at 10 perimeter locations and 9 locations further away from the facility.

The modelled vs. measured concentrations are tabulated in Table 5.12 and illustrated in Figure 5.13. The results indicate that the air model over-predicts as compared to measured concentrations, generally by a factor of 2 to 3.

Soil water is not routinely measured at Bruce Power; however, rain water is a reasonable surrogate measurement. Rain water is collected and measured at the perimeter locations. The soil water was modelled as described in Section 4.1.1 (Equation 4.1). The modelled and measured values are compared in Table 5.13 and illustrated in Figure 5.14. The results indicate that 6 out of 9 modelled values (67%) are within a factor of 2 of measured values. On average, model predictions are 57% greater than measurements.

Table 5.12: Modelled and Measured Tritium in Air at BN, 2006

Location	HTO Concentration (Bq/m³)			Model / Measured
	A/P[1]	Measured[2]	Modelled	
B2	A	2.21	7.17	3.25
B3	A	2.03	4.27	2.10
B4	A	2.09	4.13	1.98
B5	P	1.48	4.93	3.33
B6	A	0.14	0.29	2.11
B7	A	1.0	3.08	3.08
B8	A	0.21	0.39	1.85
B9	A	0.3	0.43	1.44
B10	A	1.36	3.89	2.86
B11	A	0.54	1.19	2.20
BR1	P	1.74	6.32	3.63
BR11	P	2.58	6.26	2.43
BR25	P	1.1	3.40	3.09
BR27	P	1.66	3.29	1.98
BR39	P	0.99	2.41	2.43
BF1	P	0.68	1.97	2.90
BF14	P	1.23	3.75	3.05
BDF11	P	0.25	0.65	2.58
BM6	P	0.41	0.76	1.86

[1] *A=active, P=passive, largest value was used*

[2] *Measured values background corrected, except for B6*

Table 5.13: Modelled Tritium in Soil Water Compared to Measured Tritium in Rain Water at BN, 2006

Location	HTO Concentration (Bq/L)		Model / Measured
	Rain	Modelled Soil Water	
B2	156	326.12	2.09
B4	146.5	187.86	1.28
B5	122.4	224.21	1.83
B6	6.5	13.41	2.06
B7	70.2	140.04	1.99
B8	21.5	17.61	0.82
B9	22.6	19.70	0.87
B10	69.3	176.84	2.55
B11	91.7	54.10	0.59

Figure 5.13: Modelled vs Measured Tritium in Air at BN, 2906

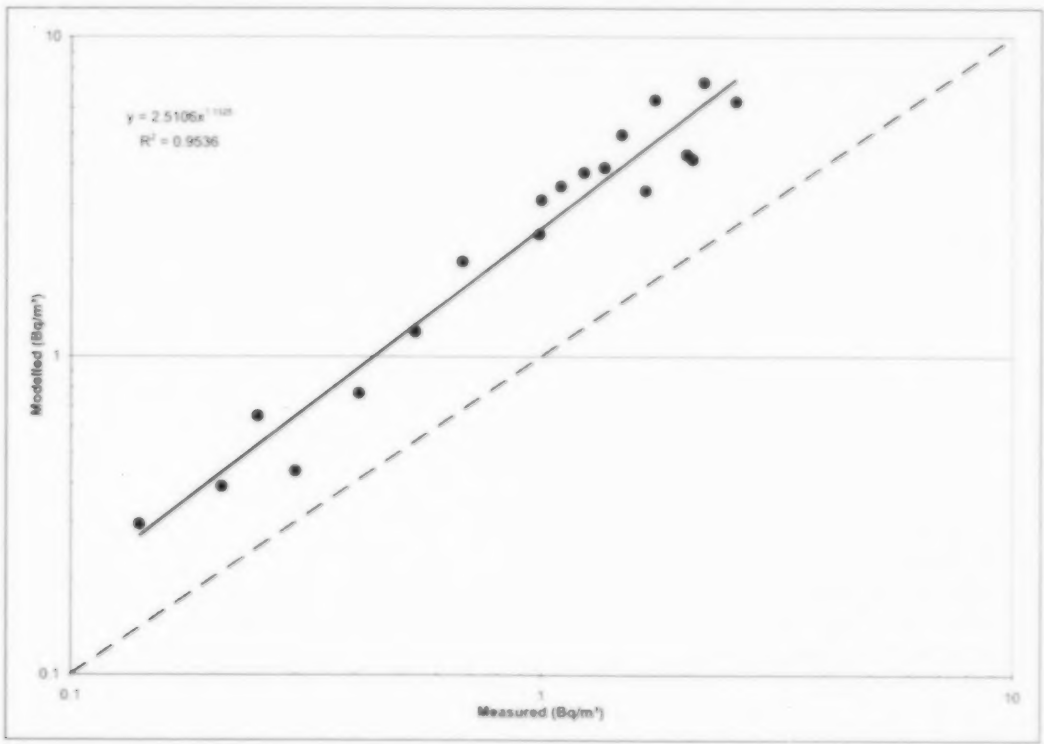
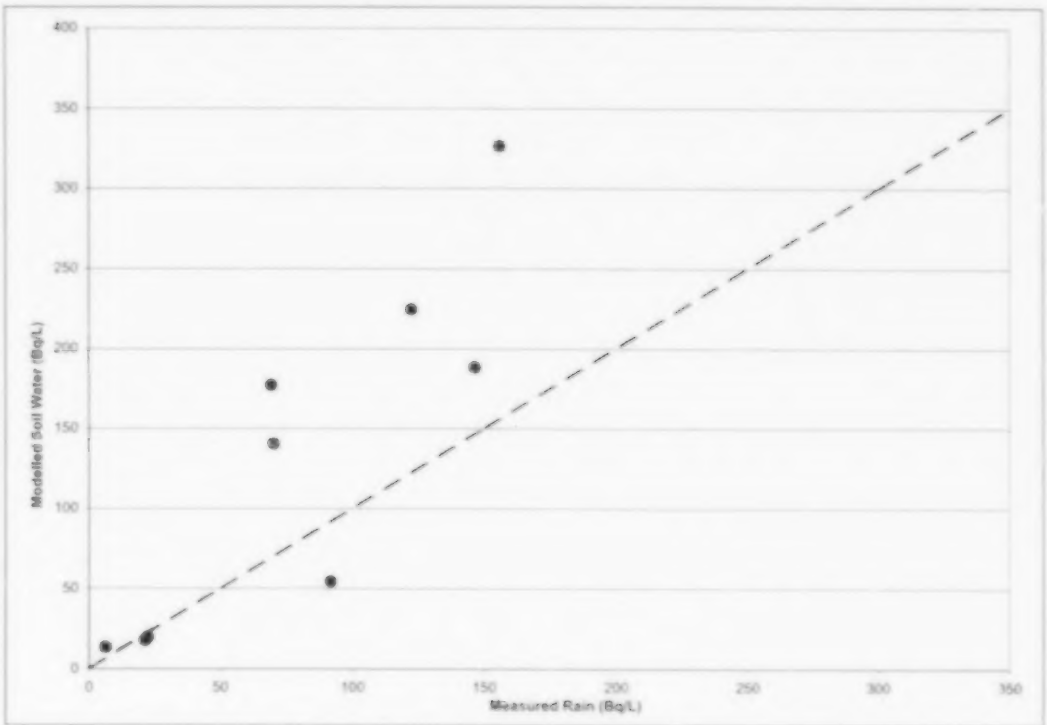


Figure 5.14: Modelled Tritium in Soil Water vs Measured Tritium in Rain Water at BN, 2006



5.3 Review of Model Performance

The foregoing examples are reviewed here in order to draw general conclusions about our ability to predict the environmental fate of tritium using simple models of atmospheric dispersion and partitioning to water. The models and parameters used are intended to be somewhat conservative, to ensure that environmental exposures to tritium are not systematically under-estimated.

5.3.1 Air

Model predictions of tritium in air were compared to measurements at five facilities known to release tritium to the atmosphere. At three facilities, the majority of predictions were within a factor of 2 of measured values, and were slightly higher than measured values on average (20-83%). At two facilities, tritium concentrations in air were over-predicted more substantially (2 or 3 fold on average). In one of these cases, the over-predictions were particularly evident in the near-field, and may stem from not representing thermal plume rise. In the other case the meteorological data represented a 5-year timeframe, rather than specifically representing the year of monitoring.

Overall, the steady-state Gaussian air model performs much as expected, generally over predicting air concentrations, more so at some facilities than others.

5.3.2 Soil Water

Model predictions of tritium in soil water were compared to measurements of soil water at one facility, and to measurements of rain water at three other facilities. Surficial soil water is expected to be similar to rain water since it originates as rain water, with a contribution from snowmelt in the spring. However, deeper soil layers may represent older rainfall, which becomes important when tritium in air is strongly increasing or decreasing.

The soil water measurements made at a facility with strongly decreasing tritium releases agreed well with soil water predictions based on the previous year's air concentrations. The majority of those predictions were within a factor of 2 of measured values, and were 35% higher on average.

The comparisons of modelled soil water to measured rain water at three facilities with relatively steady releases, showed the majority of soil water predictions within a factor of 2 of measured rain water. The predictions were approximately 60% higher on average at two facilities and approximately 2-fold higher on average at the other facility.

Overall, soil water predictions, like the air predictions on which they are based, are generally within a factor of 2 of measurements, and tend to be higher than measured values on average.

5.3.3 Pond Water

Model predictions of pond water were compared to measurements of pond water at one facility. The majority of model predictions were within a factor of 2 or 3 of measured concentrations. The model predictions averaged 28% higher than measured values. Ponds and marshes are potentially influenced by up-gradient inflow, which may provide dilution or carry tritium loading, reflecting previous inputs to the watershed. The simple model used here assumes that any flow through the pond is sufficiently small that air and pond water can equilibrate. In general, deviations from simple model predictions may be related to up-gradient hydrology.

5.3.4 Groundwater

Model predictions of groundwater at depth were compared to groundwater measurements at one facility. The majority of model predictions were within a factor of 3 of the measured concentrations. Of the predictions outside this range, one value was low as compared to measurements, likely due to localized influence of snowmelt due to snow storage in this area. Other predictions were high as compared to measurements, likely due to horizontal flow of groundwater along the bedrock surface.

In general, groundwater systems are complex as compared to a simple model of vertical transport in the aquifer. Horizontal flow, as well as local variation in sub-surface soil texture, hydraulic conductivity and infiltration rate, all can result in deviations from simple model predictions.

6.0 CONCLUSIONS AND RECOMMENDATIONS

6.1 General Conclusions

This review has presented our understanding of tritium behaviour and environmental fate in the atmospheric and hydrological environments. Specific findings on these topics are detailed in the preceding report sections. Some general conclusions are outlined below.

- Tritium behaviour in air and water is reasonably well understood. This includes dispersion in air and water, partitioning from air to soil water and surface water, and partitioning from soil water to groundwater.
- Long-term average Gaussian air models are somewhat conservative (overpredicting) and are suitable for use in estimation of annual dose to people. For modelling of shorter-term events or releases in complex terrain more sophisticated models are available.
- There are measurement uncertainties around the discrepancies between active and passive air sampler results. At some facilities, passive sampler results tend to be approximately two-fold higher.
- Air concentrations drive tritium in soil water and groundwater below a long-term atmospheric plume, assuming no nearby upgradient release to groundwater.
- Lag times can be important in soil water and groundwater since these media take months to years to equilibrate with an atmospheric plume. The lag in response of these media is particularly evident when there is a long-term trend in air concentrations. Lag times depend on substrate texture and vertical travel times.
- Groundwater flow from up-gradient, along with partitioning from the air above a well, can substantially influence tritium concentrations in well water.
- The greatest regulatory concerns related to tritium are in areas near a long-term atmospheric source (within 1 km) where people use well water and garden produce.

6.2 Recommendations

The following recommendations focus on resolving uncertainties that could influence dose estimates and safety assessments in some situations.

- Resolve the discrepancies that are often seen between active and passive air sampler results, thus reducing uncertainty in the measured values.
- Conduct near-field studies of air, soil water and groundwater designed to better understand the time lags in soil water and groundwater, and the importance of up-gradient effects.

7.0 REFERENCES

Abrol, V. 1990. Estimation of washout of tritiated water (HTO) effluent by raindrops. Bull. Rad. Prot. 13:23-66

Albenesius, E. L. 1959. Tritium as a Product of Fission, Phys. Rev. Lett., 3(6): 274-275.

Albenesius, E. L. and R. S. Ondrejcin. 1960. Nuclear Fission Produces Tritium, Nucleonics, 18(9): 100.

ANL (Argonne National Laboratory). 2005. Tritium (hydrogen-3). EVS Human Health Fact Sheet.

ATSDR (Agency for Toxic Substances and Disease Registry). 2002. Health consultation: Tritium releases and potential offsite exposures. Federal Facilities Assessment Branch, Division of Health Assessment and Consultation, Agency for Toxic Substances and Disease Registry. http://www.atsdr.cdc.gov/hac/pha/livermore2/liv_toc.html.

Audi, G. and A.H.Wapstra. 1995. The 1995 update to the atomic mass evaluation by, Nuclear Physics A595(4): 409-480.

Auld, H., J. Klaassen, S. Fernandez, G. Fenton, S. Eng, N. Comer and T. Ralph. (undated). The changing climate and NBCC climatic design values. Report for the Canadian Commission on Building and Fire Codes. Climatic Loads Task Force.

Baran, N., J. Richert, C. Mouvet. 1997. Field data and modeling of water and nitrate movement through deep unsaturated loess. Journal of Hydrology 345: 27-37.

Barrett, E.W. and L. Huebner. 1960. Atmospheric Tritium Analysis, Technical Progress Report No. 2, USAEC Report AECU-4739, University of Chicago.

Barrett, E.W. and L. Huebner. 1961. Atmospheric Tritium Analysis. Technical Progress Report No. 3, USAEC Report TID-14425, University of Chicago.

Barry, P.J. 1964. Some recent experiments on transfer of radioactive tracers from air to natural surfaces. AECL Report, AECL-2045, Chalk River, Ontario.

Baver, L. D. 1956. Soil Physics, John Wiley & Sons, Inc., New York.

Begemann, F. 1958. New Measurements on the World-Wide Distribution of Natural and Artificially Produced Tritium, in Proceedings of the Second United Nations International Conference on Peaceful Atom of Atomic Energy, Geneva. 18: 545-550.

Begemann, F. 1962. The Natural Tritium Economy of the Earth and the Question of Its Variation with Time, Chimia, 16(1): 1-10.

Begemann, F. 1963. *Earth Science and Meteorites*, North Holland Publishing Co., Amsterdam pp. 169-187.

Begemann, F. and W. F. Libby. 1957. Continental Water Balance, Ground Water Inventory and Storage Times, Surface Ocean Mixing Rates and Worldwide Water Circulation Patterns from Cosmic Ray and Bomb Tritium, Geochim. Cosmochim. Acta, 12(4): 277-296.

Bennett, B.G. 1978. Environmental aspects of americium. EML-348.

Bibron, R. 1963. Physique De L'Atmosphere, Evolution de la Concentration du Tritium dans les eaux de Precipitation en France, Compt. Rend., 256: 4951-4954.

Bibron, R. 1965. Detection of Atmospheric Tritium by Scintillation: Variations in Its Concentration in France, French Report CEA-R-2629.

Bigeleisen, J., 1962. Correlation of Tritium and Deuterium Isotope Effects. In: *Tritium in the Physical and Biological Sciences*, Symposium Proceedings, Vienna, 1961, I: 161-168, International Atomic Energy Agency, Vienna, 1962 (STI/PUB/39).

Bieringer, J and C. Schlosser. 2004. Monitoring ground-level air for trace analysis: methods and results. Anal. Bioanal. Chem., 379: 234-241.

Bishop, K.F., Delafield, H. I., Eggleton, A. E. J., Peabody, C. O., and Taylor, B. T. 1962. The Tritium Content of Atmospheric Methane. In: *Tritium in the Physical and Biological Sciences*, Symposium Proceedings, Vienna, 1961, I: 55-67, International Atomic Energy Agency, Vienna, (STI/PUB/39).

Blomeke, J.O. 1964. Management of Fission Product Tritium in Fuel Processing Wastes, USAEC Report ORNL-TM-851, Oak Ridge National Laboratory.

Bogen, D.C., G.A. Welford and C.G. White. 1979. Tritium distribution in man and his environment. In: Behaviour of Tritium in the Environment. IAEA, Vienna, p. 567-574.

Bolin, B. 1958. On the Use of Tritium as a Tracer for Water in Nature. In: *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy*. Geneva, 1958, 18: 336-343, United Nations, New York.

Bolin, E., 1961. An Investigation of Tritium in Atmospheric Moisture, Rainwater and the Sea in the European Area, IJSAEC Report TID-13376.

Bolin, B. 1964. *Research in Geophysics*, 2: 479-508. The M.I.T. Press: Cambridge, Mass.

Born, M., H. Dörr and I. Levin. 1990. Methane consumption in aerated soils of the temperate zone. Tellus, 42:2-8.

Borromeo, J. 2007. 2006 Results of Radiological Environmental Monitoring Programs. N-REP-03481-10005-R00.

Borromeo. J.. 2008. 2007 Results of Radiological Environmental Monitoring Programs. N-REP.03481-1006-R00. Ontario Power Generation.

Bowen, V.T. and W. Roether. 1973. Vertical distributions of strontium-90, caesium-137 and tritium near 45° north in the Atlantic. J. Geophys. Res., 78: 6277-6285.

Brown, R. M. and W. E. Grummitt. 1956. The Determination of Tritium in Natural Waters, Can. J. Chem., 34: 220-226.

Briggs, G.A. 1971. Plume rise: A recent critical review. Nuclear Safety 12, 15-24.

Brown, R. M. 1964. Tritium in precipitation at Canadian sites, 1953-1963. In: *Tritium and Other Environmental Isotopes in the Hydrological Cycle*, Technical Report Series, TRS No. 73, International Atomic Energy Agency, Vienna (STI/DOC/10/73).

Brown, R. M., G. L. Ogram and F. S. Spencer. 1988. Field studies of HT behaviour in the environment. 1: Dispersion and oxidation in the atmosphere. Fusion Technol., 14: 1165-1169.

Brown, T. 2005. Annual Summary and Assessment of Radiological Data for 2004. B-REP-03419-00003 R00. Bruce Power.

Bunnenberg, C. Feinhals, J., and Wiener, B. (1986). Differences in the Behaviour of HTO and H_2O in Soil after Condensation from the Atmosphere and Conversion of HT to HTO and OBT in Soil Relative to Moisture Content and Pore Volume. Rad. Protect. Dosim. 16 No. 1-2: 83-87.

Bunnenberg, C., M. Taschner, and G.L. Ogram. 1992. Key processes and input parameters for environmental tritium models. Canadian Fusion Fuels Technology Project Report CFFTP G-9032/NET-88-176, Toronto, Ontario.

Burger, L.L. 1979. Distribution and reactions of tritiated hydrogen and methane. In: Behaviour of Tritium in the Environment. IAEA, Vienna. p. 47-64.

Butler, H.L. 1963. Tritium Hazards in Heavy-Water-Moderated Reactors, Nucl. Safety, 4(3): 77-82.

Casaletto, G.J., L.H. Gevantman, and J.B. Nash. 1962. The Self-Radiation Oxidation of Tritium in Oxygen and Air. Report USNRDLTR-565, Naval Radiological Defense Laboratory.

Chamberlain, A. C. and A. E. J. Eggleton. 1964. Washout of Tritiated Water Vapour by Rain. Int J. Air Water Pollut., 8: 135-149.

Chouhan, S.L. and P. A. Davis (2001). Testing the Atmospheric Dispersion Model of CSA N288.1 with Site-Specific Data. AECL-12099. Atomic Energy of Canada Limited.

CNSC. 2003. Radioactive Release Data from Canadian Nuclear Generation Stations 1993-2002. INFO-210 (Rev 11). Canadian Nuclear Safety Commission.

Comps, F. and R.J. Doda. 1979. Large-scale distribution of tritium in a commercial production. In: Behaviour of Tritium in the Environment. IAEA, Vienna, p. 93-99.

Coyle, P.E. 1979. Laser fusion. Status, future and tritium control. In: Behaviour of Tritium in the Environment. IAEA, Vienna. p. 139-153.

Craig, H. 1957. Distribution, Production Rate, and Possible Solar Origin of Natural Tritium. Phys. Rev., 105: 1125-1127.

Craig, H. and D. Lal. 1961. The Production Rate of Natural Tritium. Tellus, 13: 85-105.

Crawford-Brown, D.J. 1984. An age-dependent model of tritium metabolism following mixed (organic/inorganic) intakes. Health Phys., 46: 924-8.

Crowson, D.L. 1973. Man-made tritium. In: Tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics, Las Vegas, Nevada p. 23-27.

Crutzen, P.J. 1991. Methane's sinks and sources. Nature 350:380-381.

CSA (2008). Guidelines for calculating derived release limits for radioactive material in airborne and liquid effluents for normal operation of nuclear facilities. Canadian Standards Association N288.1-08.

Currie, L. A. 1959. Tritium Production by 6-Bev Protons. Phys. Rev., 11413: 878-880.

Currie L. A., W. F. Libby, and R. L. Wolfgang. 1956. Tritium Production by High-Energy Protons, Phys. Rev., 101(5): 1557-1563.

Davis, P.A., 1997. Tritium transfer parameters for the winter environment. J. Environ. Rad., 36(2-3): 177-196.

Davis, P.A. and G.A. Bickel. 2000. Transfer parameters for environmental pathways models for chronic emissions of tritiated hydrogen gas. Atomic Energy of Canada Limited (AECL), RC-2501.

Davis, P.A., W.J.G. Workman, B.D. Amiro, F.S. Spencer, H. Noguchi, H. Amano, Y. Ichimasa and M. Ichimasa. 1995a. Overview of the 1994 chronic HT release experiment at Chalk River. Fusion Technology, 28: 840-845.

Davis, P.A., D.C. Galeriu, F.S. Spencer and B.D. Amiro. 1995b. Evolution of HTO concentrations in soil, vegetation and air during an experimental chronic HT release. Fusion Technology, 28: 833-839.

Davis, P., F. S. Spencer and B.D. Amiro. 1997. Evolution of HTO concentrations in soil, vegetation and air during an experimental chronic HT release. Fusion Technol. 28: 833-839.

Davis, P.A. 1995. Tritium transfer parameters for the winter environment. J. Environ. Radioactivity 36: 177-196.

Davis, P.A. 2006. Results of the EMRAS Pickering Scenario. COG Technical Report. COG-05-3070.

Davis, P.A., T.G. Kotzer and W.J.G. Workman. 2002. Environmental tritium concentrations due to continuous atmospheric sources. Fusion Science and Technology 41: 453.

DeWaele, C. 2006. Radiological Environmental Monitoring in 2005 at Chalk River Laboratories. Annual Program Report, CRL-50924-ASR-2005 (AECL-MISC-362-05-CRL, Vol 3) Rev 0. Atomic Energy of Canada Limited, Chalk River, Ontario.

Dorfman, L. M. and A. Hemmer. 1954. Ion Pair Yield of the Tritium -Oxygen Reaction. J. Chem Phys., 22: 1555-1558.

Dunstall, T.G., G.L. Ogram and F.S. Spencer. 1985a. Elemental tritium deposition and conversion in the terrestrial environment. Canadian Fusion Fuels Technology Project Report CFFTP-G-85035.

Dunstall, T.G., G.L. Ogram and F.S. Spencer. 1985b. Elemental tritium deposition and conversion in the terrestrial environment. Fusion Technol., 8: 2551-2556.

EcoMetrix. 2008. Comprehensive Report – Groundwater Studies at the SRB Technologies Facility, Pembroke, ON.

Ehhalt, D.H. 1971. Vertical profiles and transport of HTO in the troposphere. J. Geophys. Res., 76(30): 7351-7367.

Ehhalt, D.H. 1974. The atmospheric cycle of methane. Tellus, 26: 58-70.

Ehhalt, D.H., F. Rohrer, S. Schauffler and W. Pollock. 2002. Tritiated water vapor in the stratosphere: Vertical profiles and residence time. Journal of Geophysical Research, 107(D24): 4757.

Ehhalt, D.H., F. Rohrer and A. Fried. 2005. Vertical profiles of HDO/H_20 in the troposphere. Journal of Geophysical Research, 110: D13301.

Eisenbud, M. and T.F. Gesell. 1997. Environmental Radioactivity from Natural, Industrial, and Military Sources, 4th ed. Academic Press: San Diego, CA.

Engelke, M.J. and E.A. Hemis, Jr. 1962. A Study of the Diffusion and Mixing of Tritium Gas in Air, USAEC Report LA-2671, Los Alamos Scientific Laboratory.

Environment Canada. 1983. Historical Streamflow Summary. Ontario Inland Waters Directorate, Water Resources Branch, Water Survey of Canada, Ottawa.

Environment Canada. 1998. Extreme Weather and Climate Change. Report by D. Francis (Lanark House Communications) and H. Hengeveld (Atmospheric Environment Service). Minister of Supply and Services Canada.

Eriksson, E. 1965. An account of the major pulses of tritium and their effects in the atmosphere. Tellus, 17: 118-130.

Estournel, R. 1962. Presence de tritium dans les Circuits de refroidissement des reacteurs G2 et G3, French Report CEA-2205.

Faltings, V. and P. Harteck. 1950. Der tritiumgehalt der atmosphäre z. naturforschg. 5 a: 438-439.

Fireman, E.L. 1953. Measurement of the (n,H^3) cross section in nitrogen and its relationship to the tritium produced in the atmosphere. Phys. Rev., 91: 922-926.

Fireman, E. L. and F. S. Rowland. 1961. An additional measurement of the tritium content of atmospheric hydrogen of 1949. J. Geophys. Res., 66(12): 4321.

Flamm, E., R. E. Lingenfelter, J. F MacDonald, and W. F.Libby. 1962. Tritium and helium-3 in solar flares and loss of helium from the earth's atmosphere. Science, 138(3536): 48-49.

Freyer, H.D., 1977. Atmospheric cycles of trace gases containing carbon. *In: SCOPE 13 - The Global Carbon Cycle.* Workshop on the Carbon Cycle, Ratzeburg, Federal Republic of Germany, 21-26 March 1977.

Friedlander, G., J.W. Kennedy, E.S. Macias and J.M. Miller. 1981. *Nuclear and Radiochemistry*, 3rd Edition. Wiley Publishing: New York.

Freeze, R.A. and J.A. Cherry. 1979. Groundwater. Prentice-Hall, Englewood Cliffs, New Jersey. 604 pp.

Gat, J.R., U. Karfunkel, and A. Nir. 1962. Tritium content of rainwater from the eastern Mediterranean Area. In: Tritium in the Physical and Biological Sciences, Symposium Proceedings. Vienna. Vol. 1, pp. 41-54, International Atomic Energy Agency, Vienna, 1962 (STI/PUB/39).

Galeriu, D., R. Heling and A. Melintese, 2005. The dynamics of tritium including OBT in the aquatic food chain. Fusion Sci. Technol., 48(1): 779-82.

Galeriu , D., Davis, P., Raskob, W. and Melintescu, A. (2007). Tritium Radioecology and Dosimetry – Today and Tomorrow. 8th International Conference on Tritium Science and Technology. September 16 – 21, 2007, Rochester, New York.

General Nuclear Engineering Corp. 1959. A Study of Reactor Systems for the Production of Tritium and Radioactive Cobalt, USAEC Report GNEC-98.

Grosse, A.V., A.D. Kirshenbaum, J.L. Kulp, and W.S. Broecker. 1954. The natural tritium content of atmospheric hydrogen, Phys. Rev., 93: 250.

Grosse, A.V., W.M. Johnston, R.L. Wolfgang and W.F. Libby. 1951. Tritium in nature. Science, 113: 1-2.

Gratwohl, G. 1973. Erzeugung und Freisetzung von Tritium durch Reaktoren und Wiederaufarbeitungsanlagen und die voraussichtliche radiologische Belastung bis zum Jahr 2000. Kernforschungszentrum Karlsruhe report KFF-Ext. 4/73-36.

Gulden, W., W. Raskob and O. Edlund. 1990. Doses to the public due to accidental tritium releases. Fusion Technology 17: 223-227.

Gulden, W. and W. Raskob. 1992. Accidental tritium doses based on realistic modeling. Fusion Technology 21: 536-543.

Häfele, W., J.P. Holdren, G. Kessler and G.L. Kulcinski. 1977. Fusion and fast breeder reactors. IIASA RR-77-8.

Hagemann F., J. Gray Jr., L. Machta, and A. Turkevich. 1959. Stratospheric carbon-14, carbon dioxide, and tritium. Science, 130: 542-552.

Haney, W.A. 1964. Fission-product tritium in fuel-processing waste. Nucl. Safety, 4: 399-402.

Haney, W.A., D.J. Brown, and A.E. Keisenauer. 1962. Fission Product Tritium in Separations Wastes and in the Ground Water, USAEC Report IN-74536, Hanford Atomic Products Operation.

Hart, D. 2008. Derived release limits guidance. COG Technical Report, COG-06-3090.

Harteck, P. 1954. The relative abundance of HT and HTO in the atmosphere. J. Chem Phys., 22: 1746-1751.

Harteck, P. and V.F. Faltings. 1950. The helium-3 problem of the atmosphere. Nature, 166: 1109.

Hauglustaine, D.A., and D.H. Ehhalt. 2002. A three-dimensional model of molecular hydrogen in the troposphere. J. Geophys. Res., 107(D17): 4330.

Holmquist, C.E., 1965. Hazards of Tritium Oxides induced in large heavy Water Reactor Stations. Proceedings of the American Industrial Hygiene Conference, Houston, Texas. (CONF-650540-6).

Horrocks, D.L. 1964. Tritium produced during spontaneous fission of californium-252. Phys. Rev., 134: B1219.

Huber, A.H. (1984). Evaluation of a Method for Estimating Pollution Concentrations Downwind of Influencing Buildings. Atmospheric Environment 18: 2313-2338.

IAEA/WMO (International Atomic Energy Agency/World Meteorological Organization). 2004a. Management of waste containing tritium and carbon-14. IAEA Technical Reports Series No. 421.

IAEA/WMO (International Atomic Energy Agency/World Meteorological Organization). 2004b. Global network of isotopes in precipitation. The GNIP database. Available at: http://isohis.iaea.org.

ICRU (International Commission on Radiological Units and Measurements), 1963. National Bureau of Standards, Handbook 86, p. viii, Superintendent of Documents, U. S. Government Printing Office, Washington.

International Atomic Energy Agency (IAEA). 2003. Reference Biospheres for Solid Radioactive Waste Disposal: Report of BIOMASS Theme 1 of the BIOsphere Modelling and ASSessment (BIOMASS) Programme. IAEA-BIOMASS-6. Vienna.

International Lake Environment Committee (ILEC). 1999. World Lakes Database. Lake Ontario. www.ilec.or.jp/database/nam/nam-07.html.

IPCC, (2007). Climate Change 2007: Synthesis Report. An Assessment of the Intergovernmental Panel on Climate Change.

Israel, G., W. Roether and G. Schumann, 1963. Seasonal variations of bomb-produced tritium in rain, J. Geophys. Res., 68: 3771-3773.

Jackson, R.F. 1964. Experience of Operation and Use of the UK High-Flux Research Reactors, in Proceedings of the Third International Conference of the Peaceful Uses of Atomic Energy, Geneva, 7: 168-175, United Nations, New York.

Jacobs, D.G. 1968. Sources of Tritium and its Behaviour upon Release to the Environment. AEC Critical Review Series. TID-24635.

Jensen, M.R., J.F. Sykes, K.K. Tsui and P.D. Sevens-Guille. 1995. BNPD RWO Site 2; Numerical Groundwater Flow System Analysis. Nuclear Waste and Environmental Services Division, Ontario Hydro Report No. NK37-03480-94012 (VFMED) R01.

Johnson, E. F. 1962. Thermonuclear power reactors. Chem. Eng. Prop., 58: 60-61.

Jones, W.M. 1948. Thermodynamic functions for tritium and tritium hydride. The equilibrium of tritium and hydrogen with tritium hydride. The dissociation of tritium and tritium hydride. J. Chem. Phys., 16: 1077-1081.

Jones, G.C. 2007. PWR tritium issues. 8th International Conference on Tritium Science and Technology. 16-21 September 2007. Rochester, New York.

Kahn, B., R.L. Blanchard, W.L. Brinck, H.L. Krieger and H.E. Kolde. 1974. Radiological surveillance study at the Haddam Neck PWR nuclear power station. EPA-520/374-007, Washington.

Kalinowski, Martin B. and L.C. Colschen. 1995. International control of tritium to prevent horizontal proliferation and to foster nuclear disarmament. Science and Global Security. 5: 131-203.

Kaufman, S. and W. F. Libby. 1954. The natural distribution of tritium. Phys. Rev., 93: 1337-1344.

Kelley, J.H. and D. R. Tilley. 1987. TUNL nuclear data evaluation project. Nuclear Physics A474, 1.

Kim, S.B. *et al.*, 2004. Fate of Radioactive Methane Released from CANDU Reactors. COG-02-3028. February.

Kinsey, R.R. 1999. The NUDAT/PCNUDAT Program for Nuclear Data, Proceedings of the 9th International Symposium of Capture-Gamma Ray Spectroscopy and Related Topics, Budapest, Hungary. Data extracted from NUDAT database Jan. 14/1999.

Klukas, M.H. 1999. Tritium in the Great Lakes: Concentration-Time Model. Atomic Energy of Canada Ltd. (AECL) Report RC-2338.

Konig, L.A., K.G. Langguth, Pagliosa, G. Papadopoulos, A. Radziwill, A. Schulte and S. Strack. 1984. Radiookologische Studien der Auswirkungen von Tritiumemissionen am Beispiel des KfK. Report KfK 3715, Forschungszentrum Karlsruhe, Karlsruhe, Germany.

Kouts, H. and J. Long. 1973. Tritium production in nuclear reactors in tritium. (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics. Las Vegas, Nevada. p. 38-45.

Krejci, K. 1979. Discussion. In: *Behaviour of Tritium in the Environment*. IAEA, Vienna, p.101.

Krejci, K. and A. Zeller, Jr. 1979. Tritium pollution in the Swiss luminous compound industry. In: *Behaviour of Tritium in the Environment*. IAEA, Vienna, p. 65-77.

Lal, D. and B. Peters. 1967. Cosmic ray produced radioactivity on the earth. In Encyclopaedia of Physics, Vol. XLV1/2 on Cosmic Rays (K. Sitte, ed.). Springer-Verlag, New York, p. 551-612.

LaMarre, J.R. 2000. Annual Summary and Assessment of Environmental Radiological Data for 1999. Ontario Power Generation, Report N-REP-03419-10000.

Lange, N.A, and G. M. Forker. 1961. Handbook of Chemistry, 10th ed., McGraw-Hill Book Company, Inc.: New York.

Lawrence, M.G. 2005. The relationship between relative humidity and the dew point temperature in moist air: A simple conversion and applications. Bull. Am. Meteorol. Soc. 86: 225-233.

Lechnick, W. 1962. Tritium concentration in PWR using lithium-7 resin, USAEC Report WAPD-PWR-TE-103, Westinghouse Electric Corporation.

Leipunsky, O.I., 1960. Radiation hazards from clean hydrogen bomb and fission atomic bomb explosions, in fallout from nuclear weapons tests, hearings before the special.

Libby, W. F. 1952. Natural Tritium Assay, Routine Method for Absolute Assay of Beta Radioactivity, arid the Reactions of Negative π Mesons with Elementary Bromine: Final Report, USAEC file No. NP-4654.

Libby, W.F. 1953. Tritium in nature. Phys. Rev., 91: 442.

Libby, W.F. 1958. Radioactive fallout. Proc. Nut. Acad. Sci. U. S., 44: 800-820.

Libby, W.F. 1963. Moratorium tritium geophysics. J. Geophys. Res., 68: 4485-4494.

Lin, C.C. 1996. Radiochemistry in nuclear power reactors. National Academy of Sciences: Washington.

Lin, R. and K. Wei. 2005. Tritium profiles in pore water in the Chinese loess unsaturated zone: Implications for estimation of groundwater recharge. Journal of Hydrology 328: 192-199.

Locante, J. and D.D. Malinowski. 1973. Tritium in pressurized water reactors in tritium. (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics. Las Vegas, Nevada. p. 45-57.

Long, G., J. F. Cordova and S. Tanrikulu (2004). An Analysis of AERMOD Sensitivity to Input Parameters in the San Francisco Bay Area. 13th Confernce on the Applications of Air Pollution Meteorology with the Air and Waste Management Assoc.

Lucas, L.L. and M.P. Unterweger. 2000. Comprehensive review and critical evaluation of the half-life of tritium. J. Res. Natl. Inst. Stand. Technol., 105: 541-549.

MacCracken, M, (2008). Prospects for Future Climate Change and the Reasons for Early Action. J. of the Air & Waste Management Assoc. Vol. 58, 735-786.

Magee, C. B., J. E. Fulenwider, G. J. Rotariu, and R. P. Petersen. 1960. A Study of the Feasibility and Economics of Radioisotope Production in Power Reactors, Final Report, USAEC Report AECU-4355, Booz-Allen Applied Research, Inc.

Mancinelli, R.L., 1995. The regulation of methane oxidation in soil. Ann. Rev. Microbiol., 49: 581-605.

Martell, E.A. 1959. Artificial Radioactivity from Nuclear Tests up to November 1958. GRD Res. Notes No. 19, Report AFCRN-TN-5 9-444, Air Force Cambridge Research Laboratories.

Martell, E. A. 1963. On the inventory of artificial tritium and its occurrence in atmospheric methane, J. Geophys. Res., 68: 3759-3769.

Matic-Vukmirovic, Z. 1965. Determination of the tritium buildup in a heavy-water-moderated reactor. In: Proceedings of the Third International Conference on the Peaceful Uses of Atomic Energy. 3: 407-413. Geneva, United Nations, New York.

McCubbin, D., K.S. Leonard, T.A. Bailey, J. Williams and P. Tossell. 2001. Incorporation of organic tritium (^3H) by marine organisms and sediment in the Severn Estuary/Bristol Channel (UK). Marine Pollution Bulletin, 42: 852-863.

McDougall, N. 2007. Annual Summary and Assessment of Environmental Radiological Data for 2006. B-REP-03419-0007. Bruce Power.

McFarlane, J.C., R.D. Rodgers and D.V. Bradley Jr. 1978. Environmental tritium oxidation in surface soil. Environ. Sci. Technol., 12: 590-593.

McFarlane, J.C., R.D. Rodgers and D.V. Bradley Jr. 1979. Tritium oxidation in surface soils. A survey of soils near five fuel reprocessing plants. Environ. Sci. Technol., 13: 607-608.

Miskel, J.A. 1964. Characteristics of radioactivity produced by nuclear explosives. In: *Engineering with Nuclear Explosives*. Proceedings of the third plowshare symposium, Livermore, California. pp. 153-160, (TID-7695).

Miskel, J.A. 1973. Production of tritium by nuclear weapons. In: *Tritium* (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics. Phoenix and Las Vegas. p. 79-85.

Mishima, J. and C. M. Steelee (2002). Oxidation of Tritium Gas under Accident and Transport Conditions. DOE Report. LA-UR-02-3803.

Mitchel, L. 1976. Tritium inventories of the world oceans and their implications. Nature 263:103-106.

Miyamoto, K., K. Kimura and S. Hongo. 1995. A transfer model of tritium in a local hydrosphere. Fusion Technol. 28: 910-917.

Moeller, T., 1954. *Inorganic Chemistry*. John Wiley & Sons, Inc.: New York.

Momoshima, N., M. Inoue, Y. Nakamura, T. Kaji, and Y. Takashima. 1986. Recent environmental tritium levels in Japan-II coastal seawater and lake water. Journal of Radioanalytical and Nuclear Chemistry 104:141-150.

Momoshima,N., T. Okai, T. Kaji, Y. Takashima, 1991. Distribution and transformation of various chemical forms of tritium in the environment. Radiochim. Acta., 54: 129.

Momoshimam B, Yamaguchi, T., Toyoshima, T., Nagao, Y., Takahashi, M., Takamura, M. and Nakamura, Y. (2007). Tritium in the Atmospheric Environment. J. Radioanal. Nucl. Chem. April, 2007.

Morgenstern, U. 2004. Tritium dating of water and ice. *New Zealand Science Review*, 61: 42-44.

Murphy, C.E. Jr. and M.M. Pendergast. 1979. Environmental transport and cycling of tritium in the vicinity of atmospheric releases. In: *Behaviour of Tritium in the Environment*. IAEA, Vienna. p. 361-372.

Murphy, C.E. Jr., L.R. Baurer, D.W. Hayes, W.L. Marter, C.C. Zeigler, D.E. Stephenson, D.D. Hoel, and D. M Hamby. 1991. Tritium in the Savannah River Site Environment. WSRC-RP-90-424-1. Revision 1, Savannah River Site, Aiken, SC.

Mutch, R.D. and J. D. Mahony. 2008. A study of tritium in municipal solid waste leachate and gas. Fusion Science and Technology 54:305-310.

NCRP. 1996. Screening Models for Releases of Radionuclides to Atmosphere, Surface Water and Ground. NCRP Report No. 123. National Council on Radiation Protection and Measurements.

NCRP. 1979. Tritium in the Environment. NCRP No. 62. National Council on Radiation Protection and Measurements.

Neil, B.C.J. 1991. An environmental pathway model for chronic emissions of tritiated hydrogen gas. Ontario Hydro Report HSD-ST-91-21.

Neil, B.C.J. 1992. Transfer factors for the environmental pathway model CEDM-HT for chronic emissions of tritiated hydrogen gas. Ontario Hydro report HSD-ST-91-45.

Newkirk, L. L. 1963. Calculation of Low-Energy Neutron Flux in the Atmosphere by the S_n Method, J. Geophys. Res., 68: 1825-1833.

Noguchi, H., S. Fukutani, S. Yokoyama and N. Kinouchi (2001). Deposition of Tritiated Water Vapour to a Water Surface in an Outdoor Field. Radiation Protection Dosimetry 93:167-172.

Novelli, P.C., P.M. Lang, K.A. Masarie, D.F. Hurst, R. Myers and J.W. Elkins. 1999. Molecular hydrogen in the troposphere: Global distribution and budget. J. Geophys. Res., 104:427-444.

Ostlund, H.G. and R.A. Fine. 1979. Oceanic distribution and transport of tritium. In: Behaviour of Tritium in the Environment. IAEA, Vienna, p. 303-314.

Pasquill, F. (1962). Atmospheric Diffusion: The Dispersion of Windborne Material from Industrial and Other Sources. D. Van Norstand Company, Ltd., London, 1962.

Patterson, C.M. 1958. Radiation Protection Considerations in a Heavy-Water-Moderated Reactor, in Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, Vol. 23, pp. 295-301, United Nations, New York.

Paunescu, N., M. Cotarica, D. Galeriu, R. Margineanu and N. Mocanu. 1999. Evaluation of environmental tritium level in preoperational period of Cernavoda CANDU Nuclear Power Plant. Journal of Radioanalytical and Nuclear Chemistry 239: 465-470.

Qiu, X., (2002). High-Resolution Mesoscale Meteorological Modelling in Southern Ontario. Ph.D. dissertation, Faculty of Earth and Space Science, York University.

Radwan, I., Z. Pietrzak-Flis and T. Wardaszko. 2001. Tritium in surface waters, tap water and in precipitation in Poland during the 1994-1999 period. Journal of Radioanalytical and Nuclear Chemistry 247: 71-77.

Roether, W. 1967. Estimating the tritium input to groundwater from wine samples: Groundwater and direct run-off contribution to Central European surface waters. In: *Isotopes in Hydrology*. International Atomic Energy Agency, Vienna, pp. 73-79.

Roether, W. 1989. On oceanic boundary conditions for tritium, on tritiugenic ^{3}H and on the tritium ^{3}He age concept in Oceanic Circulation Models: Combining Data and Dynamics. Edited by D.L.T. Anderson and J. Willebrand. Pp. 377-407. Kluwer Acad. Norwell, Mass.

Roether, W., R. Well, A. Putzka and C.Rüth. 1998. Component separation of oceanic helium. Journal of Geophysical Research 103:27.931-27,946.

Sasaki, M., H. Kimura, H. Kudou and T. Kudou. 2000. Concentration of tritium in water vapor and precipitation on Mt. Hakkoda. Journal of Radioanalytical and Nuclear Chemistry 243: 299-303.

Schell, W.R., S. Sauzay and B.R. Payne. 1974. World distribution of environmental tritium. In: Physical Behaviour of Radioactive Contaminants in the Atmosphere. IAEA/STI/PUBI3451, IAEA, Vienna, p. 374-385

Sejkora, K. (2006). Atmospheric Sources of Tritium and Potential Implication to Surface and Groundwater Monitoring Efforts. The 16th Annual RETS-REMP Workshop, Mashantucket, CT, 26-28 June, 2006.

Serot,O., C. Wagemans and J. Heyse. 2005. New results on Helium and Tritium Gas Production from Ternary Fission. International Conference on Nuclear Data for Science and Technology. AIP Conference Proceedings, 769:857-860.

Sills, D.M.L., (1998): Lake and Land Breezes in Southwestern Ontario: Observations, Analyses and Numerical Modeling. PhD dissertation. York University.

Sloth, E.N, D.L. Horrocks, E.J. Boyce and M.H. Studier. 1962. Tritium in the thermal neutron of uranium-235, J. Inorg. Nucl. Chem., 24: 337-341.

Smith, H.A. and K. R. Fitch. 1963. Determination of the separation factor for the vaporization of mixtures of protium and tritium oxides. J. Phys. Chem., 67: 920-921.

Smith, J.M. and R.S. Gilbert. 1973. Tritium experience in boiling water reactors. in tritium (A.A. Moghissi and M.W. Carter, eds.). Messenger Graphics. Las Vegas, Nevada. p. 57-68.

Spencer, F.S., L. Vereecken-Sheehan, P.A. Davis, W.J.G. Workman, M.J. St. Aubin, D. Galeriu and B.D. Amiro. 1996. 1994 HT chronic release experiment at Chalk River: Database report. Ontario Hydro Report A-NBP-96-51-CON, AECL-11736, COG-96-603-I.

Spencer, F.S. and Vereecken-Sheehan, L. (1994). HTO Concentration Relationships Between Air, Soil and Vegetation and Seasonal Dependence of HT Deposition to Soil. Ontario Hydro Technologies, Report No. A-NBP-94-36-K.

Stamoulis, K., K. Loannides, P. Kassomenos and A. Vlachogianni. 2005. Tritium concentration in rainwater samples in northwestern Greece. Fusion Science and Technology 48: 512-515.

Stead, F. W. 1963. Tritium distribution in ground water around large underground fusion explosions. Science 142: 1163-1165.

Stebbins III, A. K. 1961. Technical Analysis Report, Second Special Report on the High Altitude Sampling Program (HASP), Report DASA539B, Defense Atomic Support Agency.

Suess, H.E. 1958. The radioactivity of the atmosphere and hydrosphere. Annu. Rev. Nucl. Sci., 8: 243.

Sültenfuß, J., 2008. Tritium: tracing the hydrological cycle Tritium – the radioactive hydrogen isotope: A multi-purpose environmental tracer for the hydrological cycle. http://www.radioaktivitaet.uni-bremen.de/Blockseminar/4_Sueltenfuss.pdf.

Spencer, F.S. and L. Vereecken-Sheehan. 1994. HTO Concentration Relationships between Air, Soil and Vegetation and Seasonal Dependence of HT Deposition to Soil. Ontario Hydro Technologies Report No A-NBP-94-36-K, COG 94-239.

SSI. 2008. 2007 Environmental Monitoring Program Annual Compliance Report. Shield Source Incorporated.

Tadmor, J. 1973. Deposition of ^{85}Kr and tritium released from a nuclear fuel reprocessing plant. Health Phys. 24: 30-43.

Taylor, C. B. 1964. Tritium in Southern Hemisphere Precipitation, 1953-1964. New Zealand Report INS-R-32.

Taylor, C. B., H. A. Polach, and T. A. Rafter. 1963. Tritium Measurements in Nature. A Review of the Work of the Tritium Laboratory Institute of Nuclear Sciences, New Zealand, New Zealand Report INS-R-24.

Thompson, S. O. and 0. A. Schaeffer. 1955. Radiation induced equilibrium of hydrogen-deuterium mixtures. J. Chem. Phys 23: 759-760.

Topp, E. and E. Pattey. 1997. Soils as sources and sinks for atmospheric methane. Can. J. Soil Sci., 77: 167-178.

Transport Canada, 2004. Handbook for Civil Aviation Medical Examiners. TP 13312E. http://www.tc.gc.ca/CivilAviation/Cam/menu.htm.

Trevorrow, L.E., B.J. Kullen and R.L. Jarry and M.J. Steindler. 1974. Tritium and noble gas fission products in the nuclear fuel cycle. I. Reactors. ANL-8102.

UN (United Nations). 1960. Tritium from Nuclear Tests, A/AC.82/R.l05/Add. 1, Secretariat, United Nations.

UN (United Nations). 1977. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation. 1977 Report to the General Assembly, with annexes. United Nations sales publication no. E.77.IX.I. New York.

UN, ILO and WHO (United Nations, International Labour Organisation and World Health Organization), 1983. Environmental health criteria for selected radionuclides. Environmental Health Criteria 25. ISBN 92 4 154085 0.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). 1977. United Nations. Sources and Effects of Ionizing Radiation United Nations Scientific Committee on the Effects of Atomic Radiation 1977 Report to the General Assembly, with annexes. United Nations sales publication no. E.77.IX.I. New York.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). 2000. United Nations. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation 2000 Report to the General Assembly, with annexes. United Nations sales publication no. E.77.IX.3. New York.

Unterweger, M.P., B.M. Coursey, F.J. Schima and W.B. Mann. 1980. Preparation and Calibration of the 1978 National Bureau of Standards Tritiated-water Standards. Int. J. Appl. Radiat. Isot., 31: 611-614.

US-DOE (United States – Department of Energy), 1993. Doe Fundamentals Handbook Nuclear Physics and Reactor Theory, Volume 2 of 2. DOE-HDBK-1019/2-93.

US EPA (2003). Comparison of Regulatory Design Concentrations: AERMOD vs ISCSt3, CTDMPLUS, ISC-PRIME. U.S. Environmental Protection Agency Office of Air Quality Planning and Standards Emissions Monitoring and Analysis Division, Research Triangle Park, North Carolina. EPA-454/R-03-002.

Verzaux, P. 1952. Investigation of the collision cross-section of lithium for neutrons. J. Phys. Radium, 31:21-27.

Von Buttlar, H. 1963. Stratospheric Tritium, Geochim. Cosmochim. Acta, 27: 741-751.

Von Buttlar, H. and W. F. Libby. 1955. Natural distribution of cosmic-ray produced tritium, Part II. J. Inorg. Nucl. Chem., l: 75-91.

Walton, A., E.L. Fisher, and P.W. Krey. 1962. Studies o f Nuclear Debris in Precipitation. USAEC Report NYO-9532, New York Operations Office.

Watson, J.C. 1961. High-energy alpha particles and tritons from the spontaneous fission of californium-252. Phys. Rev. 121: 230-231.

Wegner, H.E. 1961. Additional long-range particles from the spontaneous fission of Cf-252. RUN. Am. Phys. Soc. 6: 307.

Wehner, G. 1979. Discharges of tritium to the environment from unrestricted use of consumer products containing this radionuclide. In Behaviour of Tritium in the Environment. IAEA, Vienna, p. 79-91.

Weiss, W., J. Bullacher and W. Roether. 1979a. Evidence of pulsed discharges of tritium from nuclear energy installations in Central Europe precipitation. In: Behaviour of Tritium in the Environment. IAEA-SM-232/18, pp. 17-30.

Weiss, W., W. Roether and E. Dreisigacker. 1979b. Tritium in the North Atlantic Ocean: Inventory, input and transfer into the deep water. In: Behaviour of Tritium in the Environment. IAEA-SM-232/98, pp. 315-336.

Wilson, A. T. and G. J . Ferguson. 1960. Origin of terrestrial tritium. Geochim. Cosmochim. Acta, 18: 273-277.

Wolfgang, R. 1961. Origin of high tritium content of atmospheric methane, hydrogen, and stratospheric water. Nature 192:1279-1280.

Yamazawa, H. 2001. A one-dimensional dynamical soil–atmosphere tritiated water transport model. Environmental Modelling and Software 16:739–751.

Yang, J. Y. and L. R Gevantman. 1964. Tritium beta-radiation-induced isotopic exchange in the water vapour. J. Phys Chem. 68: 3115-3119.

Yang, J. Y. and L. R Gevantman. 1960. Tritium Beta-Radiation-Induced Isotopic Exchange in the T2-H20 System. Report USNRDGTK-471. Naval Radiological Defense Laboratory.

Yook, D., K.J. Lee, Y. Lee and D. Hong. 2006. Estimation of the tritium behavior in the Pebble Type Gas Cooled Reactor for hydrogen production. Nucl. Sci. & Technol., 43(12): 1522-1529.

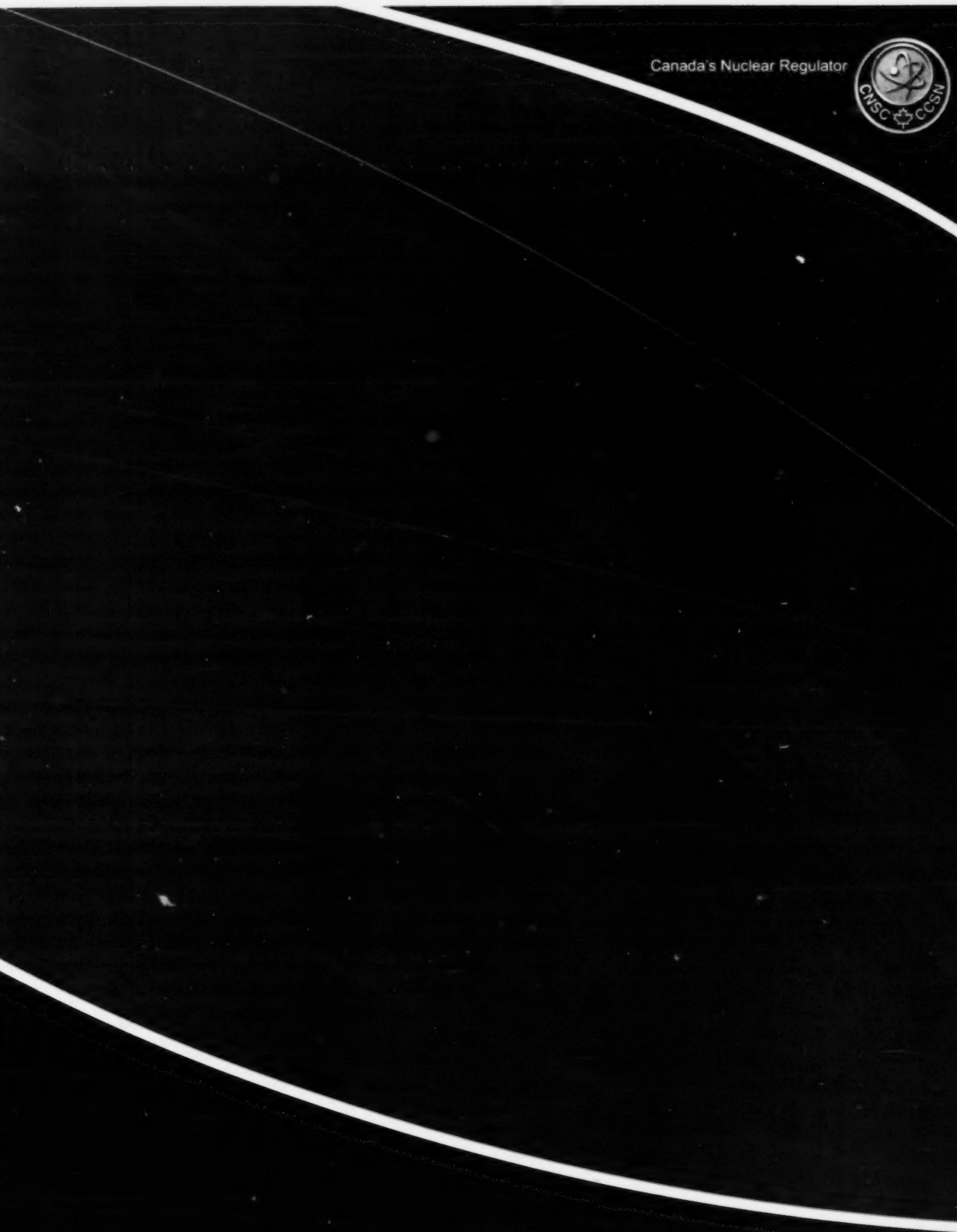
Young, P.G. and D.G. Foster, Jr. 1972. An evaluation of the neutron and gamma-ray production cross sections for nitrogen. Los Alamos Scientific Laboratory, LA-4725 (ENDF-173), UC-34.

Zahn, A., V. Barth, K. Pfeilsticker, and U. Platt. 1998. Deuterium, oxygen-18, and tritium as tracers for water vapour transport in the lower stratosphere and tropopause region. J. Atmos. Chem., 30: 25-47.

Zehnder, A.J.B. and T.D. Brock, 1979. Methane formation and methane oxidation by methanogenic bacteria. *J. of Bacteriology*, 137(1): 420-432.

Zerriffi, H., 1996. Tritium: The environmental, health, budgetary, and strategic effects of the Department of Energy's decision to produce tritium. Institute for Energy and Environmental Research (IEEE) Technical Report, http://www.ieer.org/reports/tritium.html.

Canada's Nuclear Regulator



Canadian Nuclear Safety Commission
Commission canadienne de sûreté nucléaire

Canada